Renormalized wave equation for an electron coupled to thermodynamic fluctuations of magnetization

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We derive the wave equation for an electron coupled via the s-d interaction to a system of magnetically disordered spins at nonzero temperature. A nonlinear Schrödinger equation is derived for the case of spins undergoing the Gaussian fluctuations. It shows that the character of the wave function may be significantly modified by a thermal disorder, leading to self-trapped and spin-split states in a paramagnetic medium.

The physical contents of the problem considered here are as follows. We consider an electron moving in an inhomogeneous magnetic (exchange) field created by the fluctuating spins. This field varies both in direction and in magnitude in a random way (here assumed as Gaussian). The problem then is this: what is the wave equation for the electron moving in such a random field? In other words, we are interested in deriving the Schrödinger equation in a situation in which both the direction of spin quantization axis and the magnitude of an effective magnetic field acting on the electron spin vary in a random fashion in space. To our knowledge, this problem has not been tackled this way before in the literature.

The type of problem we consider here appears, for example, when studying the bound magnetic polaron (BMP) states in a paramagnetic semiconductor. The essential role of thermodynamic fluctuations for those states has been given in detail previously.[1-3] However, the form of the electron wave function was assumed from the start and chosen based on the specific physical situation of the problem. This assumption will not be made here. We obtain a self-consistent wave equation by minimizing an appropriate form for the effective Hamiltonian[2,4] averaged with respect to spin degrees of freedom corresponding to random fluctuations.

We use the averaging procedure over random fluctuations similar to that devised previously for BMP. However, the objective of this paper is much more general. Namely, the formalism provides a method of deriving the wave equation for an arbitrary single-particle Hamiltonian H_1 and, hence, can be utilized when studying both the localized and extended stationary states.

We start from the Schrödinger equation for an electron coupled to the system of spins $\{\hat{\vec{S}}_i\}$ via the s-d interaction, $-J_c\sum_i \hat{\vec{S}}_i\cdot\hat{\vec{s}}_i$, where $\hat{\vec{s}}_i$ is the spin operator of the electron in the site representation. In the continuous version which we discuss here only, it is

$$i\hbar\partial_t\phi_\sigma(\vec{r},t)=H_1\phi_\sigma(\vec{r},t)+\tfrac{1}{2}[J_c\hat{\vec{S}}(\vec{r},t)-g^*\mu_B\vec{H}_a]\times\sum_{\sigma'}(\vec{\tau})_{\sigma\sigma'}\phi_{\sigma'}(\vec{r},t)\ , \tag{1}$$

where $\hat{\vec{S}}(\vec{r},t)=\sum_i\hat{\vec{S}}_i(t)\delta(\vec{r}-\vec{R}_n)$ is the spin-density operator. H_1 is the single-particle part of the Hamiltonian, g^* the Landé factor of the electron, $\vec{H}_a$ the applied magnetic field, and $\vec{\tau}=(\tau_x,\tau_y,\tau_z)$ the Pauli matrices. This equation contains the spin field $\hat{\vec{S}}(\vec{r},t)$, the dynamics of which is coupled to that of the electron through the s-d interaction. In what follows we calculate the reaction of the electron on itself through a response of spins to its presence. We consider two particular cases, each reflecting a different physical situation.

CASE A: SPIN-INDEPENDENT EQUATION (WEAK EXCHANGE FIELD)

The essential assumption of our approach is that the envelope function $|\phi_\sigma(\vec{r},t)|$ contains many spins.[1-3] Consequently, we assume that the adiabatic[5] and the Hartree-like[6] approximations can be used, i.e., the total wave function is of the form

$$\Psi(\{\vec{S}_i\};\vec{r},\sigma,t)=\chi\{\vec{S}_i\}\chi_\sigma\phi(\vec{r}t)\ , \tag{2}$$

where $\chi\{\vec{S}_i\}$ is the wave function of spins, and χ_σ is the spin part of that for the electron. With such a wave function one can average (1) with $\chi\{\vec{S}_i\}$ and χ_σ and get

$$i\hbar\partial_t\phi(\vec{r},t)=H_1\phi(\vec{r},t)+\frac{1}{2}\left[\frac{\alpha}{g\mu_B}\vec{M}(\vec{r})+g^*\mu_B\vec{H}_a\right]\cdot\vec{\gamma}\phi(\vec{r},t)\ , \tag{3}$$

where $\vec{M}(\vec{r})=\langle\chi|\hat{\vec{S}}(\vec{r},t)|\chi\rangle\times g\mu_B N_0$ is the local magnetization per unit volume; $N_0=n_0/v_0$ is number of atoms per unit volume containing fraction x of magnetic atoms, n_0 the number of atoms in unit cell of volume v_0, $\alpha=J_cv_0/n_0$, and $\vec{\gamma}=\sum_{\sigma,\sigma'}\langle\chi_\sigma|(\vec{\tau})_{\sigma\sigma'}|\chi_{\sigma'}\rangle$ the polarization of the electron. Under this assumption the fluctuating part of local magnetization introduces a random static potential into Eq. (3).

The procedure of averaging Eq. (3) with respect to the random part of $\vec{M}(\vec{r})$ is as follows. We first define the effective Hamiltonian[4]

$$H_{\rm eff}=\langle\phi|H_1|\phi\rangle-\tfrac{1}{2}\vec{\gamma}\cdot\vec{\Delta}[\vec{M}(\vec{r});\phi(\vec{r})]+H_s[\vec{M}(\vec{r})]\ . \tag{4}$$

The first two terms are the expectation values of the electron Hamiltonian which leads to the Schrödinger equation (3). The absolute value of the quantity

$$\vec{\Delta}[\vec{M}(\vec{r});\phi(\vec{r})]=g^*\mu_B\vec{H}_a+\frac{\alpha}{g\mu_B}\vec{M}_0(\vec{H}_a)+\frac{\alpha}{g\mu_B}\int d^3r\,[\vec{M}(\vec{r})-\vec{M}_0(\vec{H}_a)]|\phi(\vec{r})|^2 \tag{5}$$

plays a role of the spin splitting for the electron,[1,2] while $\vec{M}_0(\vec{H}_a)$ is the field-induced part of magnetization due to the spins. The last term (H_s) is the Ginzburg-Landau functional for the spins, here taken in the Gaussian form[2,7]

$$H_S[\vec{\eta}(\vec{r})]=\frac{1}{2\chi}\int d^3r\,\vec{\eta}^2(\vec{r}) \ , \tag{6}$$

where $\chi=\partial M_0/\partial H_a$ is the static susceptibility of spins, and $\vec{\eta}(r)=\vec{M}(r)-\vec{M}_0(\vec{H}_a)$.

One sees that the quantity $\vec{\Delta}/g^*\mu_B$ is an effective field acting on the electron. It contains both an applied field-induced part [the first two terms in (5)], as well as the part $\sim\vec{\eta}(\vec{r})$ which will be treated as a fluctuating field.

Next, to obtain a renormalized Hamiltonian for the electron we introduce the probability distribution of fluctuations of magnetization[7]

$$P[\vec{\eta}(\vec{r})]=\frac{\exp\{-\beta H_s[\vec{\eta}(\vec{r})]\}}{\int\mathscr{D}\vec{\eta}(\vec{r})\exp\{-\beta H_s[\vec{\eta}(\vec{r})]\}} \ , \tag{7}$$

where $\mathscr{D}\vec{\eta}(\vec{r})$ symbolizes the functional integration over all possible space profiles of $\vec{\eta}(\vec{r})$. Then, the probability distribution of electron energies $P(E_R)$, is given by

$$P(E_R)=\frac{1}{N}\sum_{\gamma=\pm1}\int\mathscr{D}\vec{\eta}(\vec{r})P[\vec{\eta}(\vec{r})] \times\exp\{-\beta(H_{\rm eff}-H_s[\vec{\eta}(\vec{r})])\} \ , \tag{8}$$

where N is the denominator of (7). Thus, the renormalized effective Hamiltonian for electron may be defined as[4,8]

$$\mathscr{H}_R=-k_BT\ln P(E_R) \ . \tag{9}$$

The functional integrations in (8) may be calculated following the method given in Ref. 2, and after some rather lengthy algebra one obtains

$$\mathscr{H}_R[\phi(\vec{r})]=\langle\phi|H_1|\phi\rangle-\tfrac{1}{2}\epsilon_p-k_BT \times\ln\left[2\cosh\left(\frac{\Delta_0}{2k_BT}\right)+\frac{4\epsilon_p}{\Delta_0}\sinh\left(\frac{\Delta_0}{2k_BT}\right)\right] \ , \tag{10}$$

where

$$\Delta_0=\frac{\alpha}{g\mu_B}M_0(H_a)+g^*\mu_BH_a \ ,$$

and

$$\epsilon_p=\frac{\alpha^2}{4(g\mu_B)^2}\int d^3r\,|\phi(\vec{r})|^4 \ .$$

This result coincides with the expressions derived before[2] for BMP, i.e., when we assume the hydrogeniclike $1s$ form of $\phi(\vec{r})$, and if H_1 is composed of the kinetic energy of the electron and of the Coulomb potential energy due to attraction by a donor. The formulation includes both bound and free magnetic polaron cases, the latter corresponding to a motion in both periodic and random potentials superposed on each other.

In order to get the renormalized wave equation for stationary states[9] we write the Euler equations for $\phi(\vec{r})$, starting from the functional $\mathscr{H}_R[\phi(\vec{r})]$, under the condition that $\int|\phi(\vec{r})|^2d^3r=1$. We have then

$$H_1\phi-\frac{1}{2}\frac{\delta\epsilon_p}{\delta\phi^*(\vec{r})}-\frac{k_BT}{\Delta_0}\frac{\delta\epsilon_p}{\delta\phi^*(\vec{r})} \times\frac{2\sinh(\Delta_0/2k_BT)}{\cosh(\Delta_0/2k_BT)+(2\epsilon_p/\Delta_0)\sinh(\Delta_0/2k_BT)}-\lambda\phi=0 \ . \tag{11}$$

It is a nonlinear differential-integral equation. The renormalized microscopic Hamiltonian $\hat{H}_R$ of the electron is defined through the relation $\hat{H}_R\phi(\vec{r})=\lambda\phi(\vec{r})$.

One of the main features of the above equation is that for extended states (Bloch states or plane waves), $\epsilon_p\sim(v_0/V)$, where V is the volume of the system. Hence, the contribution of nonlinear terms vanishes as $V\to\infty$, $(v_0/V\to0)$ for those states. This is because within a macroscopic volume, fluctuations average to zero.

The nonlinear part of (11) is attractive, so it favors localized solutions. To show this we consider the particular case of the high-temperature limit $\epsilon_p\ll k_BT$. Then, for $H_a=0$ we get

$$H_1\phi(\vec{r})-\frac{3}{4}\frac{\alpha^2\chi}{(g\mu_B)^2}|\phi(\vec{r})|^2\phi(\vec{r})=\lambda\phi(\vec{r}) \ . \tag{12}$$

The nonlinearity is stronger if χ grows. Representing $\hat{H}_R$ as $(i\hbar\partial/\partial t)$ one gets in the effective mass approximation

$$i\hbar\frac{\partial}{\partial t}\phi=-\frac{h^2}{2m^*}\nabla^2\phi-\frac{3}{4}\frac{\alpha^2\chi}{(g\mu_B)^2}|\phi|^2\phi \ . \tag{13}$$

A similar result has been derived in the mean-field approximation.[10] In one dimension, Eq. (12) has a solitary-wave solution[11] representing localized or self-trapped states:

$$\phi(x,t)=\frac{1}{\sqrt{2R}}\,\mathrm{sech}\left(\frac{x-u_et-x_0}{R}\right)\exp\left(i\frac{u_e}{2}(x-u_ct-x_0')\right) \ , \tag{14}$$

where the radius R is given by

$$R^{-1}=\frac{3}{8}\frac{\alpha^2\chi}{(g\mu_B)^2}\frac{m^*v_0^{1/3}}{\hbar^2} \ . \tag{15}$$

The expectation value for the energy using (14) is $E_e=-\hbar^2/2m^*R^2+m^*u_e^2/2$, where the first term is the gain in energy due to the nonlinear part of the potential, and the second is the kinetic energy of classical particle. For comparison, the plane-wave solution of (13), $=V^{-1/2}\times\exp[i(\vec{k}\cdot\vec{r}-E_{\vec{k}}t)]$, gives the energy

$$E_{\vec{k}}=h^2\vec{k}^2/2m^*-\tfrac{3}{4}[\alpha^2\chi/(g\mu_B)^2]V^{-1} \ .$$

The second term in $E_{\vec{k}}$ is negligible since it is proportional to (v_0/V). Therefore, in one dimension at least, the solution (14) is more favorable energetically than the extended one.

CASE B: SPIN-DEPENDENT EQUATION FOR LOCALIZED STATES

In deriving Eq. (11) we have assumed that the spin and space parts of the electron wave function can be separated [cf. Eq. (2)]. One could note, however, that for localized states, particularly for those induced by thermal disorder, the spin splitting may not be zero even though the system on the whole is not polarized magnetically.[1,2,12] In other words, the electron is localized on the fluctuation of magnetization which in turn is kept stationary by the electron field. In such a case, the wave function (2) should not be decomposed into spin and space parts, i.e., we have

$$\Psi(\{\hat{S}_i\},\vec{r}\,\vec{\gamma})=\chi\{\hat{S}_i\}\phi_{\vec{\gamma}}(\vec{r}) \ , \tag{16}$$

where $\phi_{\vec{\gamma}}(\vec{r})$ defines the wave function in locally rotated

frame in the spin space, with the z axis along the vector $\vec{\Delta}[\vec{M}(\vec{r});\phi_{\vec{\gamma}}(\vec{r})]$. This means we have now

$$\int \phi_{\vec{\gamma}}(\vec{r})\vec{\tau}\phi_{\vec{\gamma}'}(\vec{r})d^3r = \vec{\gamma}\delta_{\vec{\gamma}\vec{\gamma}'} \ .$$

Applying the same procedure as in case A, one gets the renormalized wave equation in the form (consider here only the $H=0$ case)

$$H_1\phi_\gamma(\vec{r}) - \frac{\alpha^2\chi k_B T}{2(g\mu_B)^2}|\phi_\gamma(\vec{r})|^2\frac{\delta}{\delta\epsilon_{p\gamma}}\left[\ln\left(\frac{I_1}{I_2}\right)\right]\phi_\gamma(\vec{r}) = \lambda_\gamma\phi_\gamma(\vec{r}) \ , \quad (17)$$

where $\phi_\gamma(\vec{r}) \equiv \phi_{\gamma^*}(\vec{r})$, $\gamma^* = \pm 1$,

$$\frac{I_1}{I_2} = \left(1+\frac{\epsilon_{p\gamma}}{2k_BT}\right)\exp\left(\frac{\epsilon_{p\gamma}}{2k_BT}\right)\left\{1-\Phi\left[\gamma\left(\frac{\epsilon_{p\gamma}}{2k_BT}\right)^{1/2}\right]\right\} - \frac{2\epsilon_{p\gamma}}{\pi k_BT} \ , \quad (18)$$

with $\Phi(x) = \int_0^x du\,\exp(-u^2)$ is the error function, and

$$\epsilon_{p\gamma} = \frac{\alpha^2\chi}{4(g\mu_B)^2}\int d^3r\,|\phi_\gamma(\vec{r})|^4 \ .$$

This equation gives stationary, spin-split, and self-trapped states of the electron. In the high-temperature regime ($\epsilon_{p\gamma} \ll k_BT$) one gets the renormalized Hamiltonian for one particle with spin $\gamma/2$:

$$\hat{H}_R = H_1^* - \frac{3}{4}\frac{\alpha^2\chi}{(g\mu_B)^2}\left[1-\tfrac{4}{3}\gamma\left(\frac{k_BT}{2\pi\epsilon_{p\gamma}}\right)^{1/2}\right]|\phi_\gamma(\vec{r})|^2 \ , \quad (19)$$

where H_1^* is the band term transformed to the locally rotated frame, with z axis along $\vec{\Delta}$ (cf. Ref. 13). For extended states the nonlinear part vanishes again, and we have $\hat{H}_R = H_1^*$. Each of the self-trapped states $\phi_{\gamma=-1}(\vec{r}) \equiv \phi_\downarrow$ will carry a net polarization in the paramagnetic medium. It is created by fluctuations, enhanced and sustained by the electron field. Hence, a collection of such states separated in space will give no net magnetization for $H_a = 0$.

The derivation of Eqs. (11) and (17) is our central result (the latter reduces to the former when the spin-dependent part of the nonlinear potential is disregarded). It provides a wave equation for a single electron coupled to a fluctuating field. The spin splitting is nonzero for localized states at any finite temperature, as can be seen from (18) noting that $\epsilon_p/k_BT \to 0$ continuously.

It should be stressed that the approach here describes the self-trapped states involving at least few lattice constants. However, it has been shown[14] that our theory of BMP,[1-3] which is a special case of the present approach, already gives surprisingly good results when the wave function contains four spins of magnitude $S = \frac{5}{2}$. Therefore, the additional subband structure due to the presence of spin-flip terms[6] (i.e., involving quantum corrections), and the importance of which has been shown in the atomic limit,[15] seem to be more important for rather small polarons (cf. also Ref. 6 for the case of free polaron).

The existence of the spin splitting for self-trapped states can be shown based on the following physical argument. The magnitude of the effective magnetic field acting on electron from the side of spins is of the order ($H=0$, case A)

$$H_e = \frac{\alpha}{gg^*\mu_B^2}\int d^3r\,\vec{M}(\vec{r})|\phi(\vec{r})|^2 \ .$$

This field, when averaged over all configurations of fluctuations will give

$$H_e = \frac{\alpha}{gg^*\mu_B^2}(\langle\vec{M}^2\rangle)^{1/2} \ ,$$

where $\langle\vec{M}^2\rangle$ is the mean-square amplitude of the fluctuating magnetization over the volume for which $\phi \neq 0$ (i.e., over the effective volume V_p for self-trapped states[12]). Next, from the fluctuation-dissipation theorem one can write that $\langle\vec{M}\rangle^2 = 3k_BT\chi/V_p$. Therefore, the thermodynamic average $\bar{\gamma}$ of electron polarization within the volume V_p is

$$\bar{\gamma} = \tanh\frac{|\alpha|}{2g\mu_B}\left(\frac{3\chi}{k_BTV_p}\right)^{1/2} \ .$$

It is nonzero as long as the localized solution is stable.

The eigenstates $\phi_\gamma^{(n)}(\vec{r})$ obtained from (17) provide a natural qualitative explanation of a very interesting recent observation[16] of spin-split states in the absence of applied field in semimagnetic semiconductors $Pb_{1-x}Mn_xTe$ for small $x \sim 0.01$. Namely, their spin-split states correspond to our self-trapped (free polaron) stationary states. Obviously, the quantitative analysis may require taking into account a realistic band structure, together with inclusion of higher order terms in effective spin Hamiltonian (6). Nonetheless, our method shows how to formulate the eigenvalue problem for quantum states of an electron subjected to a fluctuating exchange field.

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[4] Strictly speaking, the effective Hamiltonian $H_{\rm eff}$ here is the expectation value of a microscopic Hamiltonian. Additionally, we have neglected the denominator $\langle\phi|\phi\rangle$ in the first two terms of (4). The condition $\langle\phi|\phi\rangle = 1$ will be reinserted later [cf. Eq. (11)].

[5] The adiabatic approximation means that we calculate the electronic states for a given static and random configuration of spins. This corresponds to the Born-Oppenheimer approximation when separating electron and lattice degrees of freedom. It expresses the fact that the spins are narrowly spaced in energy (with width ΔE_s) relative to the bare bandwidth of electron states W, so that the time uncertainty $\tau_s = \hbar/\Delta E_s$ is much larger than $\tau_e = \hbar/W$. However, unlike the electron and lattice system, the amplitude of spin fluctuations is not generally small. Hence, we have to average the electron Hamiltonian over all thermally accessible spin configurations.

[6] This means that the spin-flip part of the s-d interaction does not introduce a large contribution. It is the case since we assume that there is a large number of narrowly spaced spin states with given value of $S_{\rm tot} = |\sum_i \vec{S}_i|$ within the part of space where $\phi(\vec{r}) \neq 0$. However, it means that we disregard the lifetime of scattering

states [cf. B. Sriram Shastry and D. C. Mattis, Phys. Rev. B **24**, 5340 (1981)].

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[8]Reference 7, p. 147. Equation (8) means that in the Gibbs distribution $\sim\exp(-\beta H_{\text{eff}})$ for the whole system we sum over the spin degrees of freedom. This defines a reduced probability distribution $\sim\exp(-\beta E_R)$ of electron energies $\{E_R\}$, averaged with respect to the fluctuating degrees of freedom.

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