

Nuclear spin relaxation for higher spin

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(Received 12 February 2001; published 11 May 2001)

We study the relaxation of a spin I that is weakly coupled to a quantum mechanical environment. Starting from the microscopic description, we derive a system of coupled relaxation equations within the adiabatic approximation. These are valid for arbitrary I and also for a general stationary nonequilibrium state of the environment. In the case of equilibrium, the stationary solution of the equations becomes the correct Boltzmannian equilibrium distribution for given spin I . The relaxation towards the stationary solution is characterized by a set of relaxation times, the longest of which can be shorter, by a factor of up to $2I$, than the relaxation time in the corresponding Bloch equations calculated in the standard perturbative way.

DOI: 10.1103/PhysRevB.63.224405

PACS number(s): 76.60.-k, 73.43.-f, 76.20.+q

I. INTRODUCTION

Nuclear magnetic resonance is a well-established method for testing electronic properties in solids.¹ In recent years, it became possible to apply this technique not only in three dimensions, but also to a two-dimensional electron system, the quantum Hall ferromagnet that is realized in semiconductor heterostructures in a strong magnetic field. The experimental work lead to the unexpected conclusion that a new kind of low-energy states, Skyrmions, can be formed and can determine the nuclear relaxation processes in these systems when one Landau sub-level of one spin direction is filled.²⁻¹¹

As a theoretical description of spin relaxation, Bloch's equations have been successfully used for about fifty years now. While these phenomenological equations are applicable in a wide range of cases, their microscopic derivation reveals two main restrictions. First, as was already discussed in the original work,¹² the derivation becomes strictly valid if the spin is $I=1/2$ or if the temperature of the bath is large compared to the resonance frequency. But the spin in the system under study can be $I=3/2$ (for ⁶⁹Ga, ⁷¹Ga, and ⁷⁵As; see Ref. 2), or higher in the case of magnetic impurities. Further, the progress in the experimental techniques now lets a regime of temperatures and magnetic fields come into reach, in which the temperature of the bath may be of the same order as the nuclear resonance energy (nuclear Zeeman energy). The second restriction in the derivation of the phenomenological equations demands that the environment (bath) causing the spin relaxation be in thermodynamic equilibrium. But in the case of the quantum Hall ferromagnet, the role of the bath is played by a two-dimensional electron gas in a strong magnetic field where all single particle states are degenerate into a single Landau level, and, hence, the electron-electron interaction is crucial. Such a system, dominated by Skyrmion states, is not necessarily in equilibrium. Thus, we are strongly motivated to reconsider the derivation of the Bloch equations for the relaxation of a spin, in an attempt to generalize these equations.

II. MODEL AND METHOD

In this work, we investigate the general case of (i) an arbitrary spin I and (ii) also an arbitrary stationary state of

the environment that is responsible for the relaxation. We refrain from studying a specific mechanism and consider instead the general case of a magnetic moment coupled to a bath of other quantum degrees of freedom. This magnetic moment can be a nuclear spin, or also a magnetic impurity. In the following, we use the terms "nuclear spin" for the magnetic moment and "electrons" for the bath—usually the latter is called "lattice." Then, the contribution of the nuclear spin to the Hamiltonian is

$$H = -\gamma\hat{\mathbf{I}}\cdot\mathbf{B}_0 + \hat{V}. \quad (1)$$

Below, we shall study a linear coupling between the magnetic moment $\gamma\hat{\mathbf{I}}$ (where $\hat{\mathbf{I}}$ is the spin) and an effective magnetic field generated by the electrons

$$\hat{V} = -\gamma\hat{\mathbf{I}}\cdot\hat{\mathbf{B}}. \quad (2)$$

It is well known that for $I>1/2$, there is an additional term causing relaxation, the electric quadrupole moment of the nucleus coupled to an inhomogeneous external electric field. Here, we concentrate on the linear coupling term in Eq. (2), since this model already suffices for the demonstration of our method; an inclusion of a quadrupolar coupling is straightforward. One can picture the electronic operator $\hat{\mathbf{B}}$ in Eq. (2) as being proportional to the electrons' spin. Its longitudinal component, \hat{B}_z , modifies the eigenvalues of the nuclear spin system, while the transverse components cause transitions between eigenstates. There is also a fixed part of the magnetic field in the z direction, $\mathbf{B}_0 = B_0\hat{e}_z$, which acts as an external field. The coupling between nuclear spin and electrons is supposed to be weak in the sense that we can use the adiabatic approximation as discussed below. We do not make any assumptions about the electronic subsystem's Hamiltonian or the electronic subsystem's state. This Hamiltonian may contain electron-electron interactions and the subsystem may be in an arbitrary stationary state, equilibrium or nonequilibrium.

We want to derive kinetic equations for the expectation value of the spin vector $\hat{\mathbf{I}}$ [$\hat{\mathbf{I}}^2 = I(I+1)$]. Now, for the z component, e.g., it appears one should derive, separately for each case of I , equations for the $2I+1$ diagonal elements in spinspace. But, as shall be seen below, it becomes possible to study the case of a general value of I by using spherical

tensor operators \hat{T}_{LM} as a complete basis in the space of operators acting on the state of a spin I . The \hat{T}_{LM} are irreducible tensor operators in the spherical coordinate representation,¹³ and are generally defined by their transformation properties. For actual calculations, however, the following specific definition using the spherical harmonics Y_{LM} proves very helpful:

$$\hat{T}_{LM} := \mathcal{N}_{IL} (\hat{\mathbf{I}} \cdot \nabla)^L r^L Y_{LM}(\hat{r}). \quad (3)$$

\hat{T}_{LM} is a polynomial in the components of the spin operator. It is independent of the auxiliary variable \mathbf{r} used in the definition, since there are L derivatives acting on a polynomial of order L . $\hat{r} = \mathbf{r}/r$ denotes the unit vector; we use the conventions of Ref. 13 for the spherical harmonics Y_{LM} . Then, we have $\hat{T}_{LM}^\dagger = (-1)^M \hat{T}_{L, -M}$. For a spin I , the $(2I+1)^2$ operators \hat{T}_{LM} with $L=0 \cdots 2I$ and $M=-L \cdots L$ form a complete system of operators acting in the spin space. The condition of normalization

$$Tr\{\hat{T}_{LM}^\dagger \hat{T}_{L' M'}\} = \delta_{LL'} \delta_{MM'} \quad (4)$$

determines

$$\mathcal{N}_{IL} = 2^L \sqrt{4\pi(2I-L)!/(2I+L+1)!/L!}. \quad (5)$$

Explicit expressions for the operators $\hat{T}_{L,M}$ (for $L=0 \cdots 4$) can be found in Table IV of Ref. 14.

After having established the basic notation, we proceed now to describe the derivation of the kinetic equation for the average $T_{LM}(t) = \langle \hat{T}_{LM} \rangle$, to find the stationary solution and to study finally the relaxation towards the stationary solution. Here and below, the angle brackets stand for the state of the combined system of spin and electrons. We use the framework of the Keldysh method¹⁵ in order to derive the kinetic equation. As in our earlier work on the electron spin relaxation,¹⁶ we shall employ the adiabatic approximation: the coupling between spin and electrons is supposed to be so small that in the equation of motion, its effect can be neglected beyond the first order in the spin's eigenenergies and can also be neglected beyond the second order in the relaxation times, see Ref. 17. The unperturbed motion of the spin is a precession with the frequency $\omega_0 = \gamma B_0$. Then, we have up to the second order of the perturbation theory in the coupling \hat{V} :

$$\begin{aligned} (i\partial_t - M\omega_0)T_{LM}(t) &= \langle [\hat{T}_{LM}(t), \hat{V}(t)] \rangle \\ &- i \int_{-\infty}^t dt_1 \langle [[\hat{T}_{LM}(t), \hat{V}(t)], \hat{V}(t_1)] \rangle + \mathcal{O}(\hat{V}^3). \end{aligned} \quad (6)$$

In the spirit of the adiabatic approximation, we now consider the terms of the perturbation series in higher than second order as giving rise to an additional (weak) time dependence of the spin operators due to the relaxation process, and we decouple the expectation values of spin and electron operators. The first order term $\langle [\hat{T}_{LM}, \hat{\mathbf{I}}(t)] \cdot \langle \hat{\mathbf{B}}(t) \rangle$ describes the Knight shift, the shift of the nuclear resonance frequency due

to the coupling to the electrons. Since we want to focus on the relaxation, we disregard this correction of the spin's eigenfrequency in the following. The second order term contains

$$\begin{aligned} &\langle [[\hat{T}_{LM}(t), \hat{V}(t)], \hat{V}(t_1)] \rangle \\ &= \gamma^2 \langle [\hat{T}_{LM}(t), \hat{\mathbf{I}}(t)] \cdot \hat{\mathbf{B}}(t) \hat{\mathbf{B}}(t_1) \cdot \hat{\mathbf{I}}(t_1) \\ &\quad - \hat{\mathbf{I}}(t_1) \cdot \hat{\mathbf{B}}(t_1) \hat{\mathbf{B}}(t) \cdot [\hat{T}_{LM}(t), \hat{\mathbf{I}}(t)] \rangle. \end{aligned} \quad (7)$$

Since $\hat{\mathbf{B}}$ is an operator, one gets both, commutator and anticommutator after the decoupling of the expectation values of $\hat{\mathbf{I}}$ and $\hat{\mathbf{B}}$:

$$\begin{aligned} &(i\partial_t - M\omega_0)T_{LM}(t) \\ &= -i \sum_{m; m'=0, \pm 1} \langle \langle [[\hat{T}_{LM}, \hat{I}_m], \hat{I}_{m'}](t) \rangle \rangle C_{-m; -m'} \\ &\quad + \langle \langle [[\hat{T}_{LM}, \hat{I}_m], \hat{I}_{m'}](t) \rangle \rangle R_{-m; -m'}. \end{aligned} \quad (8)$$

In deriving Eq. (8), we wrote (see the definition of $C_{m; m'}$ and $R_{m; m'}$ below)

$$\hat{I}_{m'}(t_1) \approx \hat{I}_{m'}(t) e^{im'\omega_0(t-t_1)}, \quad (9)$$

i.e., took the unperturbed time dependence of the operator $\hat{\mathbf{I}}(t_1)$ into account, but neglected in the difference $t-t_1$ in its time dependence the term arising due to the relaxation. In Eq. (8), all equal-time commutators ($[\cdot, \cdot]$) and anticommutators ($\{\cdot, \cdot\}$) can be evaluated, and the expectation values can again be expressed by $T_{LM}(t)$ as we shall see below. This remains true in the presence of a quadrupolar coupling in addition to \hat{V} ; the inclusion of such a coupling is a straightforward generalization of our approach. Thus, it is the use of the spherical tensor operators that greatly simplifies the derivation of closed coupled equations for the relaxation of the spin for arbitrary I . Most crucial are the averages of the electronic subsystem that enter Eq. (8). Since we do not assume thermodynamic equilibrium for the electronic subsystem, we get both, a correlation function and a response function that are independent, and which we denoted by $C_{m; m'}$ and $R_{m; m'}$, respectively. They are defined by

$$C_{m; m'} = \frac{\gamma^2}{2} \int_{-\infty}^0 dt' e^{im'\omega_0 t'} \langle \langle \hat{B}_m(t), \hat{B}_{m'}(t+t') \rangle \rangle, \quad (10)$$

$$R_{m; m'} = \frac{\gamma^2}{2} \int_{-\infty}^0 dt' e^{im'\omega_0 t'} \langle \langle [\hat{B}_m(t), \hat{B}_{m'}(t+t')] \rangle \rangle. \quad (11)$$

Due to the stationarity of the state of the environment, the averages are independent of the time t . $C_{m; m'}$ and $R_{m; m'}$ depend on the nuclear resonance frequency ω_0 and on the state of the environment, i.e., the temperature, if the environment is in equilibrium. Our convention for the vector components of $\hat{\mathbf{I}}$ and $\hat{\mathbf{B}}$ is $\hat{I}_{\pm 1} = \hat{I}_x \pm i\hat{I}_y$, $\hat{I}_0 = \hat{I}_z$ and $\hat{B}_{\pm 1} = (\hat{B}_x \pm i\hat{B}_y)/2$, $\hat{B}_0 = \hat{B}_z$.

Commutators and anticommutators in Eq. (8) can be calculated either directly from the definition Eq. (3), or with the aid of the theory of spherical tensors (see Ref. 18). The an-

ticommutators can be expanded in \hat{T}_{LM} ; specializing the general result containing Racah coefficients [see Eq. (17) of Ref. 18] to our case, we get for $m=0, \pm 1$:

$$\{\hat{T}_{LM}, \hat{I}_m\} = a_{L;\pm 1;M}^{(m)} \hat{T}_{L+1, M+m} + b_{L;\pm 1;M}^{(m)} \hat{T}_{L-1, M+m} \quad (12)$$

with the coefficients

$$a_{L;M}^{(0)} = b_{L;M}^{(0)} = \sqrt{L^2 - M^2} c_L, \quad (13)$$

$$a_{L;M}^{(\pm 1)} = b_{L;M\pm 1}^{(\mp 1)} = \mp \sqrt{(L+1 \pm M)(L \pm M)} c_L, \quad (14)$$

where

$$c_L = \sqrt{[(2I+1)^2 - L^2] / [(2L)^2 - 1]}. \quad (15)$$

The commutators are well-known and express the behavior of the \hat{T}_{LM} under rotations

$$[\hat{T}_{LM}, \hat{I}_m] = -d_{L;M}^{(m)} \hat{T}_{L, M+m}, \quad (16)$$

where $d_{L;M}^{(0)} = M$ and $d_{L;M}^{(\pm 1)} = \sqrt{L(L+1) - M(M \pm 1)}$.

Inserting relations (16) and (12) into Eq. (8) solves our task of deriving a closed set of relaxation equations for arbitrary nuclear spin I . The equations are linear in the expectation values of the spherical tensor operators \hat{T}_{LM} . The properties of the electronic system enter the equations parametrically in the form of correlation functions and response functions. Since the spin vector is given by the \hat{T}_{1M} , a relaxation equation for $\langle \hat{\mathbf{I}} \rangle$ can be extracted from the system of equations (8). In the case of $I=1/2$, we simply recover the Bloch equations.

In its general form, Eq. (8) is still not very transparent. Therefore, we now make additional assumptions regarding the correlation and response functions entering Eq. (8). The term $\hat{I}_z \hat{B}_z$ in the perturbation \hat{V} [corresponding to $m, m' = 0$ in Eq. (8)] just changes the spin's resonance frequency ω_0 . Since we already omitted the Knight shift, the first order correction term to this frequency, we now also omit consistently the second order contributions resulting from $m=0$ or $m'=0$ in Eq. (8). Next, the terms with $m=m'$ in Eq. (8) are neglected too. In $C_{m;m'}$ and $R_{m;m'}$, they would correspond to a twofold application of, e.g., $\hat{B}_x + i\hat{B}_y$, and that would give a total change of the z component of the electrons' spin by 2; if the electrons' state is a strict eigenstate to z component of their spin, such expectation values vanish. Under these assumptions, both correlation and response functions $C_{m;m'}$ and $R_{m;m'}$, now become "diagonal" in the index m , $C_{m;m'} = \delta_{m;-m'} C_m$ and $R_{m;m'} = \delta_{m;-m'} R_m$. Now, from the general structure of Eq. (8), it is obvious that the equations do not couple $T_{LM}(t)$ for different M . So finally, we arrive at our general result, valid for arbitrary spin I and for a nonequilibrium state of the electrons

$$\begin{aligned} & (\partial_t + iM\omega_0) T_{LM}(t) \\ &= -\frac{1}{2\tau_+} [L(L+1) - M^2] T_{LM}(t) \\ & - \frac{1}{2\tau_-} \{L\sqrt{(L+1)^2 - M^2} c_{L+1} T_{L+1, M}(t) \\ & - (L+1)\sqrt{L^2 - M^2} c_L T_{L-1, M}(t)\}. \end{aligned} \quad (17)$$

III. DISCUSSION AND CONCLUSIONS

We see that *two unrelated time scales* enter these relaxation equations

$$\frac{1}{\tau_+} := 2(C_1 + C_{-1}) \quad \text{and} \quad \frac{1}{\tau_-} := 2(R_1 - R_{-1}). \quad (18)$$

These times are independent, as long as we consider a nonequilibrium state of the electronic system. We have $\tau_+ \geq 0$. In the following, we shall frequently make use of the crucial parameter q which describes the ratio τ_+/τ_- of these times as

$$\frac{\tau_+}{\tau_-} = \frac{1-q}{1+q}, \quad (19)$$

$q \geq 0$ and $|\tau_+/\tau_-| \leq 1$. If the electrons are in equilibrium at temperature T , the fluctuation-dissipation theorem results in $q = \exp(-\omega_0/T)$.

We want to stress that the relaxation equations Eq. (17) are valid (under the assumptions stated above), no matter which specific relaxation mechanism one wants to consider. The special mechanism enters the equations in the form of *two* time scales τ_+ and τ_- . In the case of thermodynamic equilibrium in the electron system at temperature T , their ratio τ_+/τ_- is fixed by T , and only the time τ_+ —which then also depends on the temperature T —is specific for the relaxation mechanism.

Relaxation equations generally serve two purposes. First, they determine a stationary state. Second, they describe the relaxation towards this state. We now want to discuss both of these points.

Stationary solution. The study of the stationary solution of Eq. (17) serves as an important test of our procedure, since, in the case of equilibrium in the electronic system, the result is obvious. With the ansatz $T_{LM}(t) = \delta_{M,0} T_L$, one derives a recursion relation

$$c_{L+1} T_{L+1} = -\frac{\tau_-}{\tau_+} T_L + c_L T_{L-1}, \quad L=1 \dots 2I, \quad (20)$$

where $T_0 = (2I+1)^{-1/2}$, $T_{2I+1} = 0$. Then, the solution for a general nonequilibrium state of the electrons is the following stationary distribution for the z component of the spin vector

$$I_z = \sum_{m=-1}^1 m q^{-m} / \sum_{m=-1}^1 q^{-m}. \quad (21)$$

If we assume now that the electronic system is in equilibrium at temperature T , we find that I_z is given by the Brillouin function, the well-known correct equilibrium distribution for a spin I . If, on the other hand, the electronic system is driven far from equilibrium, for example into an "inverted state" where higher energies correspond to higher probabilities ($q > 1$), then that is reflected in a corresponding inverted state of the z component of the spin vector I_z .

Relaxation. The general solution of the coupled relaxation equations Eq. (17) is a superposition of exponentially decaying terms. We are mostly interested in the relaxation of the spin components $\hat{I}_z \propto \hat{T}_{10}$ and $(\hat{I}_x + i\hat{I}_y) \propto \hat{T}_{11}$, which are described by the part $M=0$ and $M=1$ of the full system of equations. For $M=0$ and $M=1$, there are $2I$ different relax-

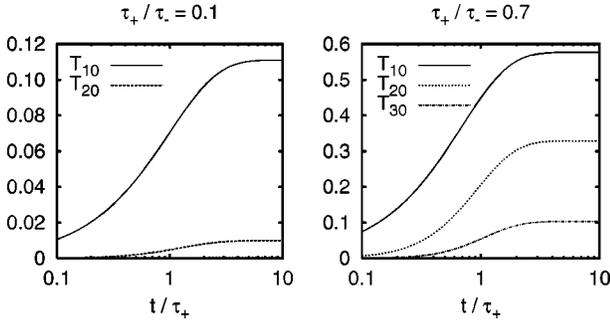


FIG. 1. Spin $I=3/2$: Solution of the relaxation equations for $M=0$ with $\tau_+/\tau_- = 0.1$ and 0.7 , respectively. The magnetization is given by $I_z(t) = \sqrt{5}T_{10}(t)$. For $\tau_+/\tau_- = 0.7$, the higher moments become significant.

ation times, for $M=2$ there are $2I-1$ times; in total we have $I(2I+3)$ times. Here, each case of I needs to be discussed separately.

In the case of spin $1/2$, Eq. (17) immediately gives the usual Bloch equations.¹ More interesting for us here, because of its experimental relevance,² is the case of spin $3/2$. The relaxation equations for $M=0$ couple the expectation values of three operators \hat{T}_{10} , \hat{T}_{20} , and \hat{T}_{30} :

$$\begin{aligned} \partial_t T_{10}(t) &= -\frac{1}{\tau_+} T_{10}(t) - \frac{1}{\tau_-} \left(\frac{2}{\sqrt{5}} T_{20}(t) - \frac{\sqrt{5}}{2} \right), \\ \partial_t T_{20}(t) &= -\frac{3}{\tau_+} T_{20}(t) - \frac{3}{\tau_-} \left(\frac{1}{\sqrt{5}} T_{30}(t) - \frac{2}{\sqrt{5}} T_{10}(t) \right), \\ \partial_t T_{30}(t) &= -\frac{6}{\tau_+} T_{30}(t) + \frac{6}{\tau_-} \frac{1}{\sqrt{5}} T_{20}(t). \end{aligned} \quad (22)$$

These equations are easily solved. In Fig. 1, we show the solution for two values of the ratio τ_+/τ_- for an initial condition of $T_{10}(0) = T_{20}(0) = T_{30}(0) = 0$ which corresponds to a completely unpolarized state of the spin. The quantities $T_{L0}(t)$ relax exponentially towards their equilibrium values. For $\tau_+/\tau_- = 0.1$, the higher moments T_{20} and T_{30} are still insignificant; $T_{30} < 0.5 \cdot 10^{-3}$ is too small to be shown in Fig. 1. On the other hand, for $\tau_+/\tau_- = 0.7$, all the moments clearly become important.

The relaxation in Eqs. (22) is described by three eigenvalues, three inverse time scales, λ , which can be simply determined from the equations as functions of q . For the case of equilibrium in the electron system, these eigenvalues of both the longitudinal ($\lambda_L, M=0$) and the transverse equations ($\lambda_T, M=1$) are shown in Fig. 2 in units of $1/\tau_+$ as functions of the temperature. Between the high-temperature limit $T \gg \omega_0$ and the low-temperature limit $T \ll \omega_0$, both the largest longitudinal and the largest transverse relaxation time decrease by a factor of three as compared to the relaxation time in Bloch approximation calculated in the usual perturbative way.

For general I , we determine both the longitudinal and the transverse relaxation times in the following limiting cases.

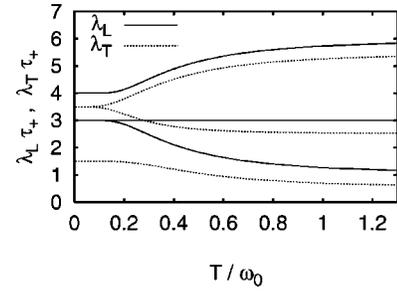


FIG. 2. Spin $I=3/2$: Inverse relaxation times in the longitudinal (solid line) and transverse (dashed line) relaxation equations in units of $1/\tau_+$.

For $\tau_+/\tau_- \sim 0$ (in case of equilibrium, $T \gg \omega_0$), the longitudinal relaxation time is τ_+ and the transverse $2\tau_+$ as in the Bloch equations. In the opposite case, $\tau_+/\tau_- \rightarrow 1$ ($T \ll \omega_0$), all the operators \hat{T}_{LM} become equally important in Eq. (17). Then, the $2I$ relaxation times are as follows ($n = 0, 1, \dots, 2I-1$): In the longitudinal equations $M=0$ we get $\tau_+ / [2(n+1)I - n(n+1)]$ and the longest relaxation time ($n=0$) is always twofold degenerate; in the transverse equations $M=1$ we get $\tau_+ / [(2n+1)I - n^2]$ and here, the longest relaxation time ($n=0$) is always nondegenerate. These explicit expressions are a conjecture based on an evaluation of the relaxation equations for spin I up to $7/2$.

Starting from the microscopic description, we have derived coupled relaxation equations valid for an arbitrary spin I in a nonequilibrium quantum environment. These equations generalize Bloch's phenomenological equations for spin relaxation. They contain two independent time scales. Their solution shows that, compared to the Bloch equations, there is an additional temperature dependence in the relaxation time which can decrease the relaxation time by a factor of up to $2I$.

These considerations allow us to draw the following conclusions for relaxation experiments. The relaxation times depend on the state of the electronic system. The electrons can be driven out of equilibrium by, e.g., optical pumping. In such a nonequilibrium case, the relaxation times T_1 and T_2 can be shorter by a factor of up to $2I$ compared with the corresponding times in the case of equilibrium. If the electronic system is in equilibrium, the experimental data must be divided by an explicit temperature dependence as it is shown in Fig. 2 when one wants to determine the relaxation time $\tau_+(T)$ which intrinsically characterizes the electronic system.

ACKNOWLEDGMENTS

We would like to thank the Deutsche Forschungsgemeinschaft for its support (Ap 47/3-1, 436 RUS 17/73/00). Yu. A.B. wishes to acknowledge support from Grants No. RFFI-00-02-17292, IR-97-0076, and INTAS 99-01146, and wishes to thank the PTB, where this work was performed, for its hospitality.

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