where in integrating over relative positions we have chosen the origin at $\mathbf{q}_1$ and the z axis is directed along the relative velocity $\mathbf{v}_2-\mathbf{v}_1$. In using cylindrical coordinates the impact parameter b is the radial coordinate which can range from zero to σ, the hard-core diameter. The azimuthal angle is denoted as ϵ. The length of the collision cylinder is $\mathbf{v}_{12}\tau_c$ so that the volume element becomes

$$d^3q_{21}=v_{12}bdbd\epsilon d\tau_c,$$

with τ_c ranging from zero to infinity. Similarly,

$$(\mathbf{k}0|e^{-\beta\Phi(q_{12})}B_z(12)|0\mathbf{k})=(z-i\mathbf{k}\cdot\mathbf{v}_1)^{-1}v_{12}\int_0^{2\pi}d\epsilon\int_0^{\sigma}bdbe^{i\mathbf{k}\cdot\boldsymbol{\sigma}(b,\epsilon)}[C(\mathbf{p}_1'\mathbf{p}_2')-C(\mathbf{p}_1\mathbf{p}_2)], \tag{B6}$$

where we have used the relation $\mathbf{q}_2=\mathbf{q}_1+(\mathbf{v}_1-\mathbf{v}_2)\tau_c(12)+\boldsymbol{\sigma}(b,\epsilon)$, with $\boldsymbol{\sigma}(b,\epsilon)$ being the vector from $\mathbf{q}_1$ to $\mathbf{q}_2$ at the instant of contact.

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Theory of Relaxation in a Group of Weakly Coupled Systems

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A general formalism for computing relaxation times characterizing two or more weakly coupled macroscopic systems is presented. The physical nature of the systems is arbitrary, although applications to spin systems are briefly discussed. The main assumptions are that each system is internally in equilibrium with a well-defined temperature, that the systems are weakly coupled to one another, and that the Hamiltonians of the systems form a commuting set. No high-temperature approximations are used. The formalism is presented in an effort to unify in part the approaches which have been developed for many special physical situations, and to show the form some relaxation times take when the high-temperature approximations are not used. It is shown that when one of two coupled systems is a "Zeeman" system, the (spin) relaxation time is proportional to the magnetic adiabatic susceptibility.

INTRODUCTION

FOR many years there has been considerable interest, experimentally and theoretically, in relaxation processes occurring in a group of two or more systems weakly coupled to one another. References 1–12 indicate some of the work concerned with these phenomena. Each of these references is concerned in some way with spin relaxation, and as a rule, treats the spin systems in a high-temperature approximation (that is, the approximation in which Curie's law is perfectly obeyed). An important exception is the work of Orbach.[8] The approach typically is to postulate certain rate equations applicable to the physical systems of interest, and from these to calculate the characteristic relaxation times. The algebra involved is quite similar in most cases.

One of our purposes in this paper is to present a unified treatment of relaxation in a group of loosely coupled systems, the physical nature of which may be left unspecified. The result is a prescription for calculating the relaxation times, namely, finding the roots of a "relaxation time matrix." Of course, general techniques for computing damping constants in electrical circuits are well known and can be found in texts on differential equations and circuit theory. However, we are not aware of any publication containing a prescription for computing relaxation times characterizing a group of macroscopic, thermal systems, and feel that this presentation fills a need.

Perhaps more important is the fact that we have avoided any high-temperature approximation. We discuss in some detail the special cases of two and three coupled systems. We show that the relaxation time characterizing two systems, one of which is a "Zeeman"

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system, is proportional to the magnetic adiabatic susceptibility.

DEVELOPMENT

We consider a group of n systems with Hamiltonians $\mathcal{H}_i$ $(i=1, 2, \cdots, n)$. Examples of the $\mathcal{H}_i$ common in spin relaxation problems are the harmonic lattice, Zeeman, spin-wave, and secular spin-spin Hamiltonians. The Hamiltonians are assumed to commute with each other. Each system is supposed to be in internal equilibrium characterized by a temperature $T_i \equiv (k\beta_i)^{-1}$, where k is Boltzmann's constant. Small interactions are assumed to exist between all pairs of systems, which eventually bring all temperatures to the same value $T \equiv (k\beta)^{-1}$. The Hamiltonians and perturbations are taken to be time-independent since we are here not interested in frequency-dependent phenomena.

Van Hove and others[13] have shown from first principles that small perturbative interactions within an isolated system (as distinguished from an open system, i.e., a system subject to external influences) bring the system into equilibrium in a manner describable by the Pauli master equation [Eq. (1) below]. The Pauli equation is a set of coupled equations for the occupation probabilities of the states of the system. The solutions to this set consist of sums of exponentially decaying terms, involving an enormous number of decay times. For more than a few degrees of freedom, the decay constants cannot be determined exactly. Although the Pauli equation undoubtedly has never been used rigorously in an analysis of the relaxation behavior of macroscopic systems, there is little doubt that it provides a suitable basis for describing, for example, physical systems for which a single relaxation time emerges as the dominating feature of the dynamics. Likewise, one expects that the Pauli equation can be used to describe a system certain portions of which come rapidly into an internal quasi-equilibrium, after which these portions come into equilibrium with each other. Most of the references given above implicitly assume that the Pauli equation, or special forms of it, can be used to describe the latter situation, and we make this hypothesis here. The principal assumption regards the form the density matrix takes [see Eq. (5)] after the subsystems have reached their internal quasi-equilibrium, namely, canonical distributions with time-varying temperatures.

The Pauli equation for the density matrix $\rho(t)$ is

$$\frac{d}{dt}\langle\phi|\rho|\phi\rangle=\sum_{\phi'} W_{\phi\phi'}[\langle\phi'|\rho|\phi'\rangle-\langle\phi|\rho|\phi\rangle]. \quad (1)$$

The state $|\phi\rangle$ is one of the complete set of eigenstates in which all $\mathcal{H}_i$ are simultaneously diagonal. Ideally, the Hilbert spaces of the different systems are mutually exclusive, so that $|\phi\rangle$ is a product state, $|\phi_1\rangle|\phi_2\rangle\cdots|\phi_n\rangle$, where $|\phi_i\rangle$ is an eigenstate of $\mathcal{H}_i$. However, there have been instances in which two commuting Hamiltonians in the same Hilbert space have been used to describe "separate" systems,[4,10] with theoretical justification[14] and experimental agreement.[10,15] To include such cases, we shall therefore make no assumption as to the separability of the states $|\phi\rangle$ into product states. $W_{\phi\phi'}$ is the transition probability per unit time, given in lowest order by

$$W_{\phi\phi'}=(2\pi/\hbar)|\langle\phi|V|\phi'\rangle|^2\delta(\mathcal{H}_\phi-\mathcal{H}_{\phi'}), \quad (2)$$

where $\mathcal{H}_\phi$ is the eigenvalue of $\mathcal{H}$ in state $|\phi\rangle$,

$$\mathcal{H}=\sum_i \mathcal{H}_i, \quad (3)$$

$$V=\sum V_{ij}, \quad (4)$$

V_{ij} is the interaction (assumed weak) between systems i and j, and the summation in Eq. (4) is over all such interactions. To account for higher order processes, V in Eq. (2) can be replaced by an "effective" interaction, as for example Orbach[8] has done in describing two-phonon processes in spin-lattice relaxation.

In accordance with the assumption that the systems are in internal equilibrium, we write

$$\langle\phi|\rho(t)|\phi\rangle=\exp(-\textstyle\sum_i\beta_i\mathcal{H}_{i\phi})/\mathrm{Tr}\{\exp(-\textstyle\sum_i\beta_i\mathcal{H}_i)\}, \quad (5)$$

where Tr is the trace operation, and the β_i are time dependent. Since $|\phi\rangle$ is not assumed to be separable, Tr likewise is not written as a product of traces.

The technique we use for generating the rate equations determining the $\beta_i(t)$ is to use Eq. (5) in the Pauli equation (1), to multiply by $\mathcal{H}_{j\phi}$, and then to sum on all states $|\phi\rangle$, thus obtaining n coupled, nonlinear differential equations in the n variables $\beta_i(t)$. The nonlinear aspects are usually, but not always,[12] avoided by the use of one or more of several techniques. These include high-temperature approximations, the assumption that one of the systems has an infinite heat capacity so that its temperature is constant, and the assumption that the temperatures are sufficiently close to the final temperature that the differential equations can be linearized in the temperature differences. For the purpose of extracting relaxation times, it is the last technique which is best, for it is in the asymptotic region of time (for which the temperature differences become small) that relaxation, as described by exponential functions, emerges from the nonlinear behavior. Further, high-temperature approximations are thereby avoided.

Since we are here interested only in computing the relaxation times, and not in the general problem of analyzing the details of the approach to equilibrium, we write

$$\beta_i(t)=\beta+\Delta\beta_i(t), \quad (6)$$

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expand the exponential functions of Eq. (5) in powers of $\Delta\beta_i$, and retain only terms linear in $\Delta\beta_i$. Thus,

$$\langle\phi|\rho(t)|\phi\rangle=\rho(\phi,\beta)[1-\sum_i \Delta\beta_i(\mathcal{H}_{i\phi}-\langle\mathcal{H}_i\rangle)], \quad (7)$$

where

$$\rho(\phi,\beta)=\exp(-\beta\mathcal{H}_\phi)/\mathrm{Tr}\{\exp(-\beta\mathcal{H})\}. \quad (8)$$

The angular brackets $\langle\ \rangle$ refer to an ensemble average with respect to $\rho(\phi,\beta)$; e.g.,

$$\langle\mathcal{H}_i\rangle=\sum_\phi \mathcal{H}_{i\phi}\rho(\phi,\beta). \quad (9)$$

Multiplying Eq. (1) by $\mathcal{H}_{j\phi}$, summing on $|\phi\rangle$, and using Eq. (7), one obtains

$$\sum_i [\langle\mathcal{H}_i\mathcal{H}_j\rangle-\langle\mathcal{H}_i\rangle\langle\mathcal{H}_j\rangle]d(\Delta\beta_i)/dt=-\sum_i \Delta\beta_i A_{ij}, \quad (10)$$

where

$$\begin{aligned}A_{ij}&=\sum_{\phi,\phi'} W_{\phi\phi'}\rho(\phi,\beta)(\mathcal{H}_{i\phi}-\mathcal{H}_{i\phi'})\mathcal{H}_{j\phi}\\&=\tfrac{1}{2}\sum_{\phi,\phi'} W_{\phi\phi'}\rho(\phi,\beta)(\mathcal{H}_{i\phi}-\mathcal{H}_{i\phi'})(\mathcal{H}_{j\phi}-\mathcal{H}_{j\phi'})\\&=A_{ji}.\end{aligned} \quad (11)$$

In deriving Eqs. (10) and (11), use has been made of the energy-conserving delta function in $W_{\phi\phi'}$. The quantity in square brackets in Eq. (10) is similar to the expression for a heat capacity. The heat capacity of system i at the final temperature T is

$$C_i=-k\beta^2\frac{\partial}{\partial\beta}\langle\mathcal{H}_i\rangle=k\beta^2[\langle\mathcal{H}_i\mathcal{H}\rangle-\langle\mathcal{H}_i\rangle\langle\mathcal{H}\rangle]. \quad (12)$$

Remember that the Hilbert spaces are not necessarily distinct. Accordingly, we shall define *partial heat capacities* C_{ij} as

$$C_{ij}\equiv k\beta^2[\langle\mathcal{H}_i\mathcal{H}_j\rangle-\langle\mathcal{H}_i\rangle\langle\mathcal{H}_j\rangle]=C_{ji}. \quad (13)$$

Equation (10) thus may be written

$$\sum_i [C_{ij}d(\Delta\beta_i)/dt+k\beta^2A_{ij}\Delta\beta_i]=0 \quad (j=1,2,\cdots,n). \quad (14)$$

Trying a solution of the form

$$\Delta\beta_i=a_ie^{-rt}, \quad (15)$$

one finds that r must satisfy the determinantal equation

$$\begin{vmatrix}k\beta^2A_{11}-rC_{11} & k\beta^2A_{12}-rC_{12}\cdots & k\beta^2A_{1n}-rC_{1n}\\ k\beta^2A_{12}-rC_{12} & k\beta^2A_{22}-rC_{22} & \\ \vdots & & k\beta^2A_{nn}-rC_{nn}\end{vmatrix}=0. \quad (16)$$

That one of the roots must be zero is seen as follows: By Eq. (11) we have

$$\sum_i A_{ij}=\sum_j A_{ij}=0. \quad (17)$$

Also, from Eqs. (12) and (13),

$$\sum_i C_{ij}=C_i, \quad (18)$$

and of course the total heat capacity C is $\sum_j C_j$. Adding each column of the determinant in Eq. (16) to the first column, and each row to the first row, one obtains

$$\begin{vmatrix}-rC & -rC_2 & -rC_3\cdots & -rC_n\\ -rC_2 & k\beta^2A_{22}-rC_{22} & k\beta^2A_{23}-rC_{23}\cdots & \\ -rC_3 & k\beta^2A_{23}-rC_{23} & & \\ \vdots & & & k\beta^2A_{nn}-rC_{nn}\end{vmatrix}=0. \quad (19)$$

Since r is a factor of each element of the first row (or column), $r=0$ is a root.

The $r=0$ root would give rise to a constant term in $\Delta\beta_i$. That this constant is zero, as it must be if the $\beta_i(t)$ eventually all acquire the same common value, is shown in the Appendix. We also show there that r must be real and non-negative.

There are thus at most $n-1$ distinct relaxation times characterizing a group of n loosely coupled systems in internal equilibrium. The inverse relaxation times are the $n-1$ roots of the determinantal equation

$$|R_{ij}|=0, \quad (20)$$

where (R_{ij}) is the $n\times n$ *relaxation time matrix*[16]

$$(R_{ij})=\begin{bmatrix}-C/r & C_2 & C_3\cdots & C_n\\ C_2 & k\beta^2A_{22}-rC_{22} & k\beta^2A_{23}-rC_{23} & \\ C_3 & k\beta^2A_{23}-rC_{23} & & \\ \vdots & & & k\beta^2A_{nn}-rC_{nn}\end{bmatrix}. \quad (21)$$

Of course, Eq. (16) could alternatively be used. Equations (20) and (21), however, are considerably more convenient.

If the Hilbert spaces of systems i and j are independent, a certain simplification of the relaxation-time matrix results. In this case $\langle\mathcal{H}_i\mathcal{H}_j\rangle=\langle\mathcal{H}_i\rangle\langle\mathcal{H}_j\rangle$, giving $C_{ij}=C_{ii}\delta_{ij}$ by Eq. (13).

TWO COUPLED SYSTEMS

When two systems are coupled, the relaxation time τ, characterizing the rate of equalization of the two tem-

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[16] This is not to be confused with the relaxation matrix of Redfield: A. G. Redfield, IBM J. Res. Develop. **1**, 19 (1957). His relaxation matrix $R_{\alpha\alpha',\beta\beta'}$ is analogous to the transition probability per unit time $W_{\phi\phi'}$.

peratures, is found from Eqs. (20) and (21) to be

$$\tau=(CC_{22}-C_2^2)/k\beta^2 AC\,, \tag{22}$$

where

$$A\equiv A_{22}=A_{11}=-A_{12}.$$

Note that

$$CC_{22}-C_2^2=C_{11}C_{22}-C_{12}^2=CC_{11}-C_1^2.$$

Equation (22) is the low-temperature generalization of formulas developed for spin-lattice[3,12] and spin-spin[10] relaxation, when applied to those systems.

Equation (22) takes a simple and physically interesting form when one of the two systems is a "Zeeman" system, whose Hamiltonian is of the form

$$\mathcal{H}_z=-MH\,, \tag{23}$$

where H is the magnitude of a magnetic field $\mathbf{H}$, and M is the magnetic-moment operator in the direction of $\mathbf{H}$. Let χ_T be the isothermal susceptibility per unit volume. Then

$$\chi_T\equiv\frac{1}{\Omega}\left(\frac{\partial\langle M\rangle}{\partial H}\right)_T=\frac{\beta}{\Omega H^2}[\langle\mathcal{H}_z^2\rangle-\langle\mathcal{H}_z\rangle^2]=\frac{C_{zz}}{k\beta\Omega H^2}\,, \tag{24}$$

where Ω is the volume. Also,

$$(\partial\langle M\rangle/\partial T)_H=-C_z/H. \tag{25}$$

The thermodynamic identity[17]

$$\chi_S=\chi_T-(T/\Omega C_H)(\partial\langle M\rangle/\partial T)_H{}^2\,, \tag{26}$$

where χ_S is the adiabatic susceptibility per unit volume, and C_H is the total heat capacity at constant H, thus yields

$$\chi_S=\frac{C_H C_{zz}-C_z^2}{k\beta\Omega H^2 C_H}\,. \tag{27}$$

Recognizing that C in Eq. (22) is the constant-field heat capacity, we see that

$$\tau=kT\chi_S\Omega H^2/A\,. \tag{28}$$

The spin-relaxation time is thus proportional to the adiabatic susceptibility when the Zeeman system is coupled to but a single additional thermal system. This simple proportionality is destroyed when two or more additional systems are involved, as will be evident from the next section. (Here "adiabatic" means that the total entropy of the set of two systems is constant.)

The expression (28), which follows from Eq. (22) without any approximations, can be written in a possibly more familiar form by considering an example.[2,12] If the "second" system is a lattice, we can write Eq. (27) as

$$k\beta\Omega H^2\chi_S=C_zC_l/(C_z+C_l)\,, \tag{27a}$$

and Eq. (28) as

$$\tau=kT^2C_z/A(1+C_z/C_l)\equiv T_1/(1+C_z/C_l)\,, \tag{28a}$$

where T_1 is the spin-lattice relaxation time for infinite lattice heat capacity. The factor $1+C_z/C_l$ directly reflects the existence of χ_S rather than χ_T in Eq. (28): This factor goes to unity, and $\chi_S\to\chi_T$, as $C_l\to\infty$. An analogous factor, $1+\mathrm{Tr}\mathcal{H}_z^2/\mathrm{Tr}\mathcal{H}_{\mathrm{sec}}^2$, where $\mathcal{H}_{\mathrm{sec}}$ is the secular spin-spin Hamiltonian, appears in the high-temperature longitudinal spin-spin relaxation time formula.[10] The definition of T_1 by Eq. (28a) is the low-temperature generalization of the usual formula[3,12] for spin-lattice relaxation. In the spin-spin case, formulas similar to Eqs. (27a) and (28a) cannot be written, except at high-temperature, because the Zeeman and secular Hamiltonians have the same Hilbert space.

THREE COUPLED SYSTEMS

In this section, we exhibit the general form of the quadratic equation from which the two relaxation times characterizing three coupled systems may be found, and give an example. Equations (20) and (21) give

$$r^2-\frac{k\beta^2}{|C_{ij}|}[A_{22}\Gamma_{33}+A_{33}\Gamma_{22}-2A_{23}\Gamma_{23}]r+\frac{k^2\beta^4C}{|C_{ij}|}(A_{22}A_{23}-A_{23}^2)=0\,, \tag{29}$$

where

$$\Gamma_{ij}=CC_{ij}-C_iC_j\,, \tag{30}$$

and $|C_{ij}|$ is the determinant of the partial heat-capacity matrix.

As an example of Eq. (29), we consider that the three systems have distinct Hilbert spaces, and that the heat capacity of system 1 is infinite. This would correspond to the situation analyzed by Schumacher,[5] in which system 1 corresponds to an infinite "lattice", and systems 2 and 3 correspond to two separate spin systems. We have

$$\frac{\Gamma_{22}}{|C_{ij}|}=\frac{1}{C_3}\,;\quad\frac{\Gamma_{33}}{|C_{ij}|}=\frac{1}{C_2}\,;\quad\frac{\Gamma_{23}}{|C_{ij}|}=-\frac{1}{C_1}\to 0. \tag{31}$$

In addition, the quantities A_{ij}, defined in Eq. (11), now separate into sums. Using

$$\langle\phi_i\phi_j\phi_k|V_{ij}|\phi_i'\phi_j'\phi_k'\rangle=\langle\phi_i\phi_j|V_{ij}|\phi_i'\phi_j'\rangle\delta_{\phi_k\phi_k'}\,, \tag{32}$$

and

$$\sum_{\phi_k}\rho(\phi_i,\phi_j,\phi_k;\beta)=\rho(\phi_i,\phi_j;\beta)\,, \tag{33}$$

we find that

$$\begin{aligned}A_{22}&=B_{12}+B_{23}\,,\\A_{33}&=B_{13}+B_{23}\,,\\A_{23}&=-B_{23}\,,\end{aligned} \tag{34}$$

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[17] Related identities may be found, for example, in the review article: A. H. Cooke, Rept. Progr. Phys. **13**, 276 (1950).

where

$$B_{ij}=\tfrac{1}{2}\sum(2\pi/\hbar)|\langle\phi_i\phi_j|V_{ij}|\phi_i'\phi_j'\rangle|^2 \times\delta(\mathcal{H}_{i\phi_i}+\mathcal{H}_{j\phi_j}-\mathcal{H}_{i\phi_i'}-\mathcal{H}_{j\phi_j'}) \rho(\phi_i,\phi_j;\beta)(\mathcal{H}_{i\phi_i}-\mathcal{H}_{i\phi_i'})^2. \quad (35)$$

With the change of notation

$$\begin{aligned} R_2&=k\beta^2B_{12}/C_2, & R_3&=k\beta^2B_{13}/C_3, \\ R_{23}&=k\beta^2B_{23}/C_2, & R_{32}&=k\beta^2B_{23}/C_3, \end{aligned} \quad (36)$$

(to facilitate comparison with Schumacher's work), we can now write Eq. (29) as

$$r^2-(R_2+R_3+R_{23}+R_{32})r+R_2R_3+R_2R_{32}+R_3R_{23}=0. \quad (37)$$

When systems 2 and 3 are taken as spin systems, and a high-temperature approximation is used for each, Eq. (37) gives Schumacher's two relaxation times.

Another well-known three-system group consists of the Zeeman energy of a set of spins coupled to the secular energy (that part of the spin-spin interaction which commutes with the Zeeman energy) of the same spins, which are in turn coupled to the lattice vibrational energy, or to the crystalline-field energy. This has been discussed by Bloembergen *et al.*[6] Fletcher, LeCraw, and Spencer,[18] and Sparks[19] have discussed three- and four-system groups involving magnons.

We conclude by mentioning that although no high-temperature approximations have been made in the formalism presented here, the formulas are not necessarily valid at arbitrarily low temperatures. A coupling mechanism which is weak at high temperature can well become strong at low temperature, as is the case with spin-spin interactions. Additional restrictions on the temperature arise when two Hamiltonians have the same Hilbert space.[14] On the other hand, the usual high-temperature approximation for spin systems is unnecessarily restrictive. Deviations of the spin heat capacity from the T^{-2} form, and deviations from Curie's law can be appreciable well within the paramagnetic region.

ACKNOWLEDGMENT

I wish to thank R. M. Wilcox for pointing out that the reality and non-negative character of the roots of the relaxation-time matrix could be demonstrated very simply.

APPENDIX

We first show that the roots r of the relaxation-time matrix must be real and non-negative. Substituting Eq. (15) into Eq. (14), multiplying by a_j^*, summing on j, and using the definitions of C_{ij} and A_{ij}, one obtains

$$r\langle|\textstyle\sum_i a_i\Delta\mathcal{H}_i|^2\rangle =\tfrac{1}{2}\sum_{\phi,\phi'}W_{\phi\phi'}\rho(\phi,\beta)|\textstyle\sum_i a_i(\mathcal{H}_{i\phi}-\mathcal{H}_{i\phi'})|^2, \quad (\mathrm{A1})$$

where

$$\Delta\mathcal{H}_i\equiv\mathcal{H}_i-\langle\mathcal{H}_i\rangle.$$

From Eq. (A1) it follows at once that r is real and non-negative. (Since all r are real, the a_i of Eq. (15) must be real to match the initial conditions.)

Any zero root of the relaxation-time matrix gives rise to a constant term in the expression for $\Delta\beta_i(t)$. That such a term must be zero is seen as follows. By the above discussion, Eq. (14) goes to

$$\textstyle\sum_i A_{ij}\Delta\beta_i(\infty)=0 \quad (j=1,2,\cdots,n), \quad (\mathrm{A2})$$

as $t\rightarrow\infty$, where $\Delta\beta_i(\infty)$ designates the possibly non-vanishing limiting value of $\Delta\beta_i(t)$. By Eq. (17), only $n-1$ equations of the set (A2) are independent. An additional equation derives from the conservation of energy of the n systems, in the form

$$\textstyle\sum_i C_i\Delta\beta_i(t)=0. \quad (\mathrm{A3})$$

Equation (20) is the standard thermodynamic expression, written to terms linear in the $\Delta\beta_i$, but also follows immediately from Eqs. (7), (8), (12), and

$$\textstyle\sum_\phi\langle\phi|\rho(t)|\phi\rangle\mathcal{H}_\phi=\sum_\phi\rho(\phi,\beta)\mathcal{H}_\phi. \quad (\mathrm{A4})$$

Equation (A4) expresses conservation of energy and follows from the Pauli equation (1). Thus there are n independent homogeneous equations, which over determine the n variables $\Delta\beta_i(\infty)$, showing that $\Delta\beta_i(\infty)=0$.

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[18] R. C. Fletcher, R. C. LeCraw, and E. G. Spencer, Phys. Rev. **117**, 955 (1960).

[19] M. Sparks, *Ferromagnetic-Relaxation Theory* (McGraw-Hill Book Company, Inc., New York, 1964).