Effect of correlated self-diffusion on the low-field nuclear-spin relaxation in the rotating reference frame

Dieter Wolf

Max-Planck-Institut für Metallforschung, Institut für Physik, Stuttgart, West-Germany

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The Slichter-Ailion (SA) theory for low-field rotating-frame relaxation by atomic diffusion in crystals is applied to a monovacancy mechanism of self-diffusion under the condition that a spin temperature is established between successive jumps of a nucleus but not between successive jumps of a vacancy (so-called high-temperature region). By means of a computer simulation of the random migration of a vacancy both the existence of a "trail of hot spins" left behind the vacancy and the correlated jumps of neighboring nuclei are taken into account. As special cases the theory includes low-temperature relaxation, where a spin temperature is established even between two successive vacancy jumps, and uncorrelated random-walk diffusion. The extrapolation to high relaxation fields shows the identity of the high-field case as predicted from the SA theory with the results obtained from a perturbation-theory treatment of relaxation due to vacancy-induced self-diffusion by the present author. From the numerical results for the fcc and bcc lattices considerable variations of the orientation dependence of the rotating-frame spin-lattice relaxation time are predicted for both the transition from low temperatures to high temperatures and for the transition from the low-field to the high-field region.

I. INTRODUCTION

Nuclear-magnetic-relaxation experiments for studying atomic motions in crystals may roughly be classified into two groups: (i) studies of the laboratory-frame spin-lattice relaxation time T_1 and the spin-spin relaxation time T_2 in the vicinity of the T_1 minimum; (ii) application of "ultraslow motion" techniques to the measurement of the spin-lattice relaxation time $T_{1\rho}$ in the rotating coordinate system in the vicinity of the $T_{1\rho}$ minimum.

While the T_1 minimum occurs whenever the mean time of stay τ of a nucleus at a lattice site is comparable with a precession period ω_0^{-1} of the spins in the strong constant external field $\vec{H}_0$ (i.e., whenever $\omega_0\tau \approx 1$), the $T_{1\rho}$ minimum occurs when $\omega_1\tau \approx 1$, where ω_1 is the Larmor precession frequency associated with the rotating field $\vec{H}_1$. Since ω_1 is much smaller than ω_0 (typically by about three to four orders of magnitude) the temperature range over which atomic motions may be detected is considerably extended to low temperatures by the application of the so-called Slichter-Ailion (SA) technique for the measurement of $T_{1\rho}$.[1,2]

In most monoatomic crystals diffusion results from randomly migrating point defects (e.g., vacancies, interstitials) by which the directions and the time sequence of successive jumps of an atom may be correlated, so that they are not independent of one another. While the activation energy of the diffusion mechanism may be simply determined from a logarithmic plot of the relaxation times versus reciprocal temperature, considerable theoretical effort is necessary to relate the measured relaxation time to the diffusion coefficient as obtained, e.g., from measurements with radioactive tracers.

Two different theoretical approaches are used to relate the mean time τ (characteristic for a given diffusion mechanism) to the relaxation times in high fields or low fields: (i) In the high-field region (i.e., for $H_0 \gg H_L$ in the laboratory frame, or $H_1 \gg H_{L\rho}$ in the rotating frame, where H_L and $H_{L\rho}$ are the local fields in the coordinate systems involved) the splitting of the Zeeman levels is large compared with the dipolar coupling of the spins. Hence between successive nuclear jumps no cross relaxation between the dipolar and Zeeman Hamiltonian occurs and the dipolar Hamiltonian may be treated as a small time-dependent perturbation on the Zeeman Hamiltonian. This procedure, comprehensively described by Abragam,[3] was recently applied to correlated and uncorrelated diffusion mechanisms in crystals by the present author.[4] (ii) In the low-field region (i.e., for $H_1 \lesssim H_{L\rho}$), the spins are strongly coupled, and the perturbation method fails. The communication between the dipolar and Zeeman Hamiltonian is now rather fast, so that cross relaxation may occur between successive nuclear jumps. Therefore, SA[1] suggested to characterize the nuclear spin system by a common spin temperature of the two spin Hamiltonians in the rotating frame. This spin temperature relaxes towards the lattice temperature as thermal equilibrium is established. After a perturbation (e.g., a jump of an atom or a point defect) the spin temperature is established within a time of the order of $T_2^{r\prime}$, the spin-spin relaxation time in a "rigid" lattice, i.e., in the absence of atomic jumps. The SA theory (see, e.g., Ref. 5) is applicable if between successive nuclear jumps the spin system may be characterized by a common spin temperature of the Zeeman

and dipolar Hamiltonians, i.e., if $T_2^{rl} < \tau$.

As pointed out earlier[6] the jumps of a given nucleus are bunched into groups if they are caused by a defect mechanism. In addition, the jumps of neighboring atoms are correlated with the jumps of that nucleus. For typical vacancy concentrations ($C_v \approx 10^{-4}-10^{-7}$), we have $\tau_v \ll \tau$, where τ_v is the mean time of stay of a vacancy at a lattice site. Then, within the framework of the SA theory we have the two regions $\tau_v \gg T_2^{rl}$ and $\tau_v \ll T_2^{rl}$ (but still $\tau > T_2^{rl}$).

If $\tau_v \gg T_2^{rl}$ (low-temperature region of the SA theory), a spin temperature is established between successive jumps of the vacancy, and the bunching effect mentioned above does not influence the relaxation process. This situation was treated in detail for different diffusion mechanisms by Ailion and Ho.[7] At temperatures, however, where the condition $\tau_v \gg T_2^{rl}$ holds, the diffusion contribution to the relaxation rate is very often dominated by motion-independent relaxation mechanisms (e.g., the conduction electron contribution in metals), so that no information on diffusion may be obtained from the measured relaxation times.

For $\tau_v \ll T_2^{rl} \ll \tau$ (high-temperature region), a spin temperature is not established between successive vacancy jumps nor between those jumps of the nuclei which are caused by the same vacancy. The relaxation process is governed by the correlated motions of the nuclei. Qualitatively, this situation was described by SA in terms of a trail of "hot" spins (which are not at the common spin temperature) left behind by the vacancy.

The present paper deals with a quantitative treatment of the relaxation process by correlated self-diffusion via monovacancies for the case $\tau_v \ll T_2^{rl} \ll \tau$.

II. APPLICATION OF ENCOUNTER MODEL

In a previous paper[4] the effect of correlated relative motions of the nuclei on high-field relaxation was described in terms of the so-called encounter model. In the following this model will be applied to low-field relaxation due to a randomly migrating vacancy.

We consider a spin i located at the origin of the coordinate system. We assume that the orientations of spin i and of the neighboring spins may be characterized by a spin temperature. When a vacancy causes atomic jumps in the surroundings of spin i it destroys the common spin temperature. By definition,[4] in an encounter with the vacancy spin i jumps on the average $Z(0)$ times, while a neighboring spin m performs $Z(\vec{r}_m^0)$ jumps (averaged over many individual encounters). When the vacancy leaves the neighborhood of spin i all spin positions have been rearranged relative to each other, and a new spin temperature is estab-
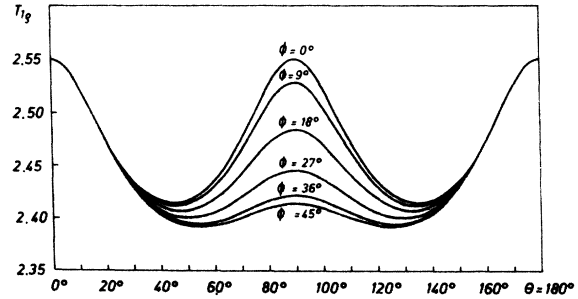

FIG. 1. Orientation dependence of $T_{1\rho}$ in zero field in units of $\tau/2$ for $0 \le \theta \le 180°$ and ϕ between 0° and 45° [see Eqs. (8.1) and (8.2) and Table I].

lished within a time of order T_2^{rl}. (Because of the condition $\tau_v \ll T_2^{rl}$, the spin orientations are assumed not to be changed during the actual rearranging procedure.)

If Δt_{NMR} denotes the mean time between two different rearrangements, the SA theory may be applied to calculate the relaxation rate, if $\Delta t_{NMR} \gg T_2^{rl}$. An estimate of Δt_{NMR} may be obtained in the following way.

From numerical calculations it is found that with an accuracy of about 2% the energy of the dipolar interactions of spin i with all other spins of the sample (i.e., the orientation of spin i) is given by its interaction with the first $n \approx 130$ to 150 nearest-neighboring spins only. Therefore, we assume that the orientation of spin i can change only if the position of one of these n neighboring spins is changed relative to spin i, and that jumps of nuclei outside the "surroundings" of spin i (defined in this way by the number n) do not contribute to the relaxation of spin i. The number of relative jumps caused by the vacancy during its encounter with spin i is given by[4]

$$Z_{NMR} = Z(0) + \sum_{m=1}^{n} Z(\vec{r}_m^0) . \tag{2.1}$$

Since Z_{NMR}/n denotes the mean number of jumps per spin in one encounter, on the average the time

$$\Delta t_{NMR} = \tau\, Z_{NMR}/n \tag{2.2}$$

elapses before a different vacancy becomes active in the surroundings of spin i. Numerically, it is found[4] that Z_{NMR} is about 40–50 for the first $n \approx 140$ spins. Thus, we obtain from Eq. (2.2) the estimate

$$\Delta t_{NMR} \approx \tfrac{1}{3}\tau . \tag{2.3}$$

According to the SA theory,[5] the spin-lattice relaxation time in the rotating frame $T_{1\rho}$ is related to the rate of change of the mean dipolar interaction energy T_c^{-1} by

$$\frac{1}{T_{1\rho}} = \frac{H_{L\rho}^2}{H_1^2 + H_{L\rho}^2} \frac{1}{T_c} . \qquad (2.4)$$

From the encounter model described above T_c may be calculated as follows. Let $\langle \Delta E_{NMR}(i, m) \rangle$ be the mean change of the mean dipolar interaction energy $\langle E_D(i, m) \rangle$ between spin i and an arbitrary spin m of the crystal, resulting from an encounter of i. Since the number of relative jumps of i and m is $Z(0) + Z(\vec{r}_m^0)$ [if spin m is not located in the "surroundings" of spin i, pair correlation is neglected (see Ref. 4), and $Z(\vec{r}_m^0) = 0$], the rate of change of $E_D(i, m)$ is $\langle \Delta E_{NMR}(i, m) \rangle / \tau_{NMR}(m)$, where[4]

$$\tau_{NMR}(m) = \tfrac{1}{2} \tau [Z(0) + Z(\vec{r}_m^0)] . \qquad (2.5)$$

The factor of 2 arises from the fact that $\langle E_D(i, m) \rangle$ changes if either of the spins i and m makes an encounter. Hence we obtain for the total relative change of the mean dipolar energy of one spin per second

$$\frac{1}{T_c} = \frac{1}{\langle E_D \rangle} \sum_m \frac{\langle \Delta E_{NMR}(i, m) \rangle}{\tau_{NMR}(m)} , \qquad (2.6)$$

where

$$\langle E_D \rangle = \sum_m \langle E_D(i, m) \rangle \qquad (2.7)$$

denotes the mean dipolar energy per spin.

III. CALCULATION OF THE ENERGY CHANGE PER ENCOUNTER

As pointed out in Ref. 4, an encounter of spin i with a vacancy is looked upon as representative for the vacancy-induced motions of all spins of the sample. Hence it is sufficient to calculate the energy change resulting from one encounter of spin i to describe the relaxation behavior of the entire sample.

The secular part $\mathcal{H}_{d_r}^0$ of the dipolar Hamiltonian of spin i in the rotating frame, $\mathcal{H}_{d_r}$, may be written (see, e.g., Ref. 5)

$$\mathcal{H}_{d_r}^0 = \tfrac{1}{4} \gamma^2 \hbar^2 \sum_m F_{im}^{(0)} (3 I_{iz} I_{mz} - \vec{I}_i \vec{I}_m) \qquad (3.1)$$

where, as in the laboratory frame,

$$F_{im}^{(0)} = r_{im}^{-3} (1 - 3 \cos^2 \theta_{im}) \qquad (3.2)$$

The assumption of a spin temperature prior to an encounter (i.e., before the vacancy causes the first jump in the surroundings of spin i) is equivalent with the assumption of the following density matrix of the spin system:

$$\sigma = (1/Z) \exp\{-[(\mathcal{H}_{d_r}^0)_b + \mathcal{H}_{z_r}]/k\theta\} , \qquad (3.3)$$

where

$$Z = (2I + 1)^N \qquad (3.4)$$

in the high-temperature approximation. $(\mathcal{H}_{d_r}^0)_b$ denotes the dipolar Hamiltonian of spin i *before* the encounter; θ is the spin temperature. For the Zeeman Hamiltonian in the rotating frame we have[5]

$$\mathcal{H}_{z_r} = \gamma \hbar \sum_n [(H_0 - \omega/\gamma) I_{n_z} + H_1 I_{n_x}] . \qquad (3.5)$$

In resonance $\omega = \omega_0 = \gamma H_0$, and the first term on the right-hand side of Eq. (3.5) vanishes. From Eqs. (3.1) and (3.3), we obtain, for the mean dipolar energy of spin i,

$$\langle E_D \rangle = \tfrac{1}{4} \gamma^4 \hbar^4 U \sum_m |(F_{im}^{(0)})_b|^2 , \qquad (3.6)$$

where U includes the traces over the spin operators. In Sec. II it was assumed that because of the condition $\tau_v \ll T_2^{r'}$ the orientation of the nuclear spins is not changed while the vacancy actually rearranges the surroundings of spin i, so that the density matrix immediately after the encounter is still given by Eq. (3.3) (so-called sudden approximation). In terms of the encounter model the result of the rearrangement is a "sudden" change of the dipolar Hamiltonian from $(\mathcal{H}_{d_r}^0)_b$ to $(\mathcal{H}_{d_r}^0)_f$, while the Zeeman Hamiltonian remains unchanged during the rearranging procedure. During the process of spin-lattice relaxation (assumed not to begin before the end of the encounter) this change of the dipolar Hamiltonian is communicated to the Zeeman Hamiltonian, and can be observed as a change of magnetization. For the expectation value $\langle \Delta E_{NMR} \rangle$ of the mean change of the dipolar Hamiltonian of spin i due to an encounter we may thus write analogously with[5]

$$\langle \Delta E_{NMR} \rangle = \mathrm{Tr}\{\sigma[(\mathcal{H}_{d_r}^0)_b - (\mathcal{H}_{d_r}^0)_f]\}$$

$$= \frac{1}{k\theta(2I+1)^N} [\mathrm{Tr}(\mathcal{H}_{d_r}^0)_b^2 - \mathrm{Tr}(\mathcal{H}_{d_r}^0)_b (\mathcal{H}_{d_r}^0)_f] . \qquad (3.7)$$

Inserting Eq. (3.1) into (3.7), we obtain

$$\langle \Delta E_{NMR} \rangle = \tfrac{1}{4} \gamma^4 \hbar^4 U \sum_m [|(F_{im}^{(0)})_b|^2 - (F_{im}^{(0)})_b (F_{im}^{(0)})_f]$$

$$= \sum_m \langle \Delta E_{NMR}(i, m) \rangle . \qquad (3.8)$$

In the rest of this section the quantities $(F_{im}^{(0)})_b$ and $(F_{im}^{(0)})_f$ will be specified in terms of the encounter model.

Analogously to Ref. 4, $B_g(\vec{r}_m^0, \vec{r}_m)$ is defined as the probability that spin m, before the encounter at $\vec{r}_m^0$ relative to spin i, will be located at the lattice site $\vec{r}_m$ after the encounter, if the first jump of i was to the nearest-neighbor site g in the crystal lattice ($g = 1, \ldots, G$; G is the number of nearest neighbors). Similarly, the probabilities $B_g(0, \vec{r}_i)$ describe to possible jumps of spin i during the encounter.

Then we may write

$$\sum_m |\,(F_{im}^{(0)})_b\,|^2 = \sum_{\vec{r}_m^0} |\,F_{im}^{(0)}(\vec{r}_m^0)\,|^2 \qquad (3.9)$$

and

$$\sum_m (F_{im}^{(0)})_b (F_{im}^{(0)})_f = \sum_{\vec{r}_m^0} F_{im}^{(0)}(\vec{r}_m^0)$$

$$\times \frac{1}{G} \sum_{g=1} \sum_{\vec{r}_i} \sum_{\vec{r}_m} B_g(0, \vec{r}_i)\, B_g(\vec{r}_m^0, \vec{r}_m)\, F_{im}^{(0)}(\vec{r}_m - \vec{r}_i) \ . \qquad (3.10)$$

In Eq. (3.10) an average was taken over the G nearest-neighboring positions in the crystal lattice, from which the vancancy may cause the first jump of spin i. The vector $\vec{r}_m - \vec{r}_i$ from i to m after the encounter may be the same discrete lattice vector for several combinations of $\vec{r}_m$ and $\vec{r}_i$. As in Ref. 4, the vector $\vec{r}_m^*$ of the effective relative displacement of the two spins is defined by the relationship

$$\vec{r}_m - \vec{r}_i = \vec{r}_m^0 + \vec{r}_m^* \ . \qquad (3.11)$$

Let $W_1(\vec{r}_m^0, \vec{r}_m^*)$ denote the probability that the relative displacement due to the encounter is $\vec{r}_m^*$, provided $\vec{r}_m^0$ is the initial vector from i to m. As shown in Ref. 4 [Eq. (4.10a)],

$$W_1(\vec{r}_m^0, \vec{r}_m^*) = \frac{1}{G} \sum_{g=1}^{G} \sum_{\vec{r}_i} B_g(0, \vec{r}_i)\, B_g(\vec{r}_m^0, \vec{r}_m^0 + \vec{r}_m^* + \vec{r}_i) \ , \qquad (3.12)$$

and from Eqs. (3.10)–(3.12), we find

$$\sum_m (F_{im}^{(0)})_b\, (F_{im}^{(0)})_f = \sum_{\vec{r}_m^0} F_{im}^{(0)}(\vec{r}_m^0)$$

$$\times \sum_{\vec{r}_m^*} W_1(\vec{r}_m^0, \vec{r}_m^*)\, F_{im}^{(0)}(\vec{r}_m^0 + \vec{r}_m^*) \ . \qquad (3.13)$$

With these results the relaxation rate T_c^{-1} of the dipolar energy may be calculated. Inserting Eqs. (3.13), (3.9), (3.8), (3.6), and (2.5) into (2.6), we obtain

$$\frac{1}{T_c} = \frac{2}{\tau \sum_{\vec{r}_m^0} |\,F_{im}^{(0)}(\vec{r}_m^0)\,|^2} \sum_{\vec{r}_m^0} \frac{|\,F_{im}^{(0)}(\vec{r}_m^0)\,|^2 - F_{im}^{(0)}(\vec{r}_m^0) \sum_{\vec{r}_m^*} W_1(\vec{r}_m^0, \vec{r}_m^*)\, F_{im}^{(0)}(\vec{r}_m^0 + \vec{r}_m^*)}{Z(0) + Z(\vec{r}_m^0)} \ . \qquad (3.14)$$

Analogously to SA, we write

$$1/T_c = 2(a - p)/\tau \ , \qquad (3.15)$$

and from Eq. (2.4),

$$1/T_{1\rho} = [H_{L\rho}^2/(H_1^2 + H_{L\rho}^2)]\, 2(a - p)/\tau \ , \qquad (3.16)$$

with

$$a = \sum_{\vec{r}_m^0} \frac{|\,F_{im}^{(0)}(\vec{r}_m^0)\,|^2}{Z(0) + Z(\vec{r}_m^0)} \Big/ \sum_{\vec{r}_m^0} |\,F_{im}^{(0)}(\vec{r}_m^0)\,|^2 \qquad (3.17)$$

and

$$p = \sum_{\vec{r}_m^0} \frac{F_{im}^{(0)}(\vec{r}_m^0) \sum_{\vec{r}_m^*} W_1(\vec{r}_m^0, \vec{r}_m^*)\, F_{im}^{(0)}(\vec{r}_m^0 + \vec{r}_m^*)}{Z(0) + Z(\vec{r}_m^0)} \Big/ \sum_{\vec{r}_m^0} |\,F_{im}(\vec{r}_m^0)\,|^2 \ . \qquad (3.18)$$

The local field $H_{L\rho}$ in the rotating frame may be calculated from the relationship[5]

$$H_{L\rho}^2 = \tfrac{1}{4} \gamma^2 \hbar^2 I(I+1) \sum_{\vec{r}_m^0} |\,F_{im}^{(0)}(\vec{r}_m^0)\,|^2 \ . \qquad (3.19)$$

Equations (3.16)–(3.18) describe the relaxation process resulting from a randomly migrating point defect with a mean time of stay at a lattice site τ_d, which is small compared with T_2^{rl}, so that correlation effects have to be taken into account. In Sec. VIII the quantities a and p will be evaluated numerically from Eqs. (3.17) and (3.18) for diffusion via monovacancies.

IV. COMPARISON WITH RESULTS OF SA

As mentioned in Sec. I, a spin temperature is established between successive vacancy jumps if $T_2^{rl} \ll \tau_v \ll \tau$ (low-temperature region), and the correlation of successive jumps of a nucleus as well as the correlation of the jumps of different nuclei (so-called pair correlation) does not affect the relaxation process.

In terms of the encounter model described above this situation may be treated as the limit in which the "encounter" consists of a single vacancy jump only. The unimportance of pair correlations is equivalent with assuming

$$B_g(\vec{r}_m^0, \vec{r}_m) = \delta_{\vec{r}_m, \vec{r}_m^0} \qquad (4.1)$$

and

$$Z(\vec{r}_m^0) = 0 ,\qquad (4.2)$$

where $\delta_{\vec{r}_m^0, \vec{r}_m}$ denotes the Kronecker symbol (equals 1 for $\vec{r}_m^0 = \vec{r}_m$, and 0 otherwise). Since the vacancy initiates a jump of spin i from one of the g nearest-neighboring sites in the crystal, we have

$$B_g(0, \vec{r}_i) = (1/G)\,\delta_{\vec{r}_i, \vec{r}_g} \qquad (4.3)$$

In the calculation of the energy change per vacancy jump in Eqs. (3.9) and (3.10) it has to be taken into account that before and after the jump of spin i a vacancy is on a nearest neighboring lattice site. If the summations over all nuclei m are replaced by summations over all lattice sites $\vec{r}_m^0$ the contribution resulting from the vacant site has to be subtracted. Thus we find, from Eqs. (3.9), (3.10), (4.1), and (4.3),

$$\sum_m |\,(F_{im}^{(0)})_b\,|^2 = \sum_{\vec{r}_m^0} |\,F_{im}^{(0)}(\vec{r}_m^0)\,|^2 - \frac{1}{G}\sum_{g=1}^{G} |\,F_{im}^{(0)}(\vec{r}_g)\,|^2 \qquad (4.4)$$

and

$$\sum_m (F_{im}^{(0)})_b\,(F_{im}^{(0)})_f = \frac{1}{G}\sum_{\vec{r}_m^0} F_{im}^{(0)}(\vec{r}_m^0) \sum_{g=1}^{G} F_{im}^{(0)}(\vec{r}_m^0 - \vec{r}_g) . \qquad (4.5)$$

Since for $\tau_v \gg T_2^{rl}$ between two jumps of spin i a new spin temperature is established [(i.e., $Z(0)$ $=1$], we obtain, in analogy to Eq. (3.14),

$$\frac{1}{T_c} = 2\left(\sum_{\vec{r}_m^0} |\,F_{im}^{(0)}(\vec{r}_m^0)\,|^2 - \frac{1}{G}\sum_{g=1}^{G} |\,F_{im}^{(0)}(\vec{r}_g)\,|^2\right.$$
$$\left. -\frac{1}{G}\sum_{\vec{r}_m^0} F_{im}^{(0)}(\vec{r}_m^0) \sum_{g=1}^{G} F_{im}^{(0)}(\vec{r}_m^0 - \vec{r}_g)\right)\Big/$$
$$\left(\tau \sum_{\vec{r}_m^0} |\,F_{im}^{(0)}(\vec{r}_m^0)\,|^2\right) \qquad (4.6)$$

or

$$1/T_c = 2(1 - p)/\tau , \qquad (4.7)$$

with

$$p = \frac{1}{G}\sum_{g=1}^{G}\left(|\,F_{im}^{(0)}(\vec{r}_g)\,|^2 + \sum_{\vec{r}_m^0} F_{im}^{(0)}(\vec{r}_m^0)\,F_{im}^{(0)}(\vec{r}_m^0 - \vec{r}_g)\right)\Big/$$
$$\sum_{\vec{r}_m^0} |\,F_{im}(\vec{r}_m^0)\,|^2 . \qquad (4.8)$$

Equations (4.7) and (4.8) were first derived by SA[1] and Ailion and Ho (Ref. 7) under the assumption that $\tau_v \gg T_2^{rl}$.

Starting from Eqs. (4.7) and (4.8), SA presented a qualitative solution to the problem of vacancy-induced relaxation in the high-temperature region (i.e., for $\tau_v \ll T_2^{rl} \ll \tau$). They corrected

Eq. (4.7) for the following two effects: (i) Since before the first jump of spin i in the encounter typically one of its nearest neighbors is hot (i.e., it is not at the common spin temperature), this spin is assumed not to contribute to the relaxation rate, and Eq. (4.7) has to be reduced by a factor of $1 - 1/G$.

Actually, the "trail of hot spins" left behind by the vacancy includes more than only one spin. In our model (Sec. III) this is accounted for by the jumps of spins in the surroundings of spin i before the first jump of i, which destroy the common spin temperature. If $B_g^b(\vec{r}_m^0, \vec{r}_m')$ denotes the probability that a spin m, initially located at $\vec{r}_m^0$, will sit at $\vec{r}_m'$ when spin i jumps the first time, then the probability $H_g(m)$, that spin m is hot at that moment, may be calculated from the relationship

$$H_g(m) = \sum_{\vec{r}_m' \neq \vec{r}_m^0} B_g^b(\vec{r}_m^0, \vec{r}_m') . \qquad (4.9)$$

(ii) Correlation effects due to the random motion of the vacancy were taken into account by SA by a further reduction of the relaxation rate (4.7) by the factor $1 - 2/G$, the first approximation for the spatial correlation factor. In the encounter model these correlations are accounted for by the probabilities $W_1(\vec{r}_m^0, \vec{r}_m^*)$ [see Eq. (3.12)].

With these two corrections of Eq. (4.7), the result of SA for the case $\tau_v \ll T_2^{rl} \ll \tau$ is usually written

$$\frac{1}{T_{1\rho}} = \frac{H_{L\rho}^2}{H_1^2 + H_{L\rho}^2}\,\frac{2(1 - p)}{\tau}\,\frac{G - 1}{G}\,\frac{G - 2}{G} . \qquad (4.10)$$

The main difference between the relaxation rate (3.16) and the approximation (4.10) is that in (4.10) the same orientation dependence of $T_{1\rho}$ for vacancy induced diffusion is predicted for both cases, $\tau_v \gg T_2^{rl}$ and $\tau_v \ll T_2^{rl}$, while the orientation dependence of $T_{1\rho}$ resulting from (3.16) is substantially different from that predicted for low temperatures [see Eqs. (4.6)–(4.8) and Table I].

V. LIMITING CASE OF UNCORRELATED RANDOM-WALK SELF-DIFFUSION

High-field nuclear magnetic relaxation resulting from random-walk self-diffusion in monoatomic crystals was treated by Torrey[8] and by the present author.[4] This model is not very realistic in that it assumes jumps to nearest-neighbor lattice sites without the presence of vacancies or interstitial atoms. Nevertheless, random-walk diffusion is of some theoretical interest, because it may be considered as a limiting case of correlated diffusion mechanisms.[4]

Similar to the case of vacancy-induced diffusion in the low-temperature region described in See. IV, we assume a spin temperature to be established between successive jumps of the nuclei

TABLE I. Numerical values for single crystals and for polycrystalline samples for monovacancy diffusion in the high-temperature region (present theory and approximation of Allion and Ho) and in the low-temperature region (Ailion and Ho). The results for a random-walk diffusion model shown here may be obtained from the values published by Ailion and Ho (Ref. 7) for monovacancy diffusion. $[\delta = \frac{3}{8}\gamma^2\hbar^2 I(I+1)$, $2a_0 =$ cube edge of a unit cell.]

		Present model $[\tau_v \ll T_2^{\tau l} \ll \tau$, see (7.7)]	Ailion and Ho (Ref. 7) $[\tau_v \ll T^{\tau l} \ll \tau$, see (4.10)]	Ailion and Ho (Ref. 7) $[\tau_v \gg T_2^{\tau l}$, see Eq. (4.7)]	Random walk $[\tau \gg T_2^{\tau l}$, see Eq. (5.1)]
A in units of a_0^{-6}	bcc	0.0911	0.1100	0.1676	0.1676
	fcc	0.4719	0.6592	0.8623	0.9248
B in units of a_0^{-6}	bcc	0.0726	0.0656	0.1000	0.1440
	fcc	0.2058	0.2569	0.3363	0.3832
$a - p$ [see Eq. (8.2)]	bcc	$[0.0911 + 0.0726 f(\theta,\phi)]$ $[0.2324 + 0.1622 f(\theta,\phi)]$	$[0.1100 + 0.0656 f(\theta,\phi)]$ $[0.2324 - 0.1622 f(\theta,\phi)]$	$[0.1676 + 0.1000 f(\theta,\phi)]$ $[0.2324 + 0.1622 f(\theta,\phi)]$	$[0.1676 + 0.1440 f(\theta,\phi)]$ $[0.2324 + 0.1622 f(\theta,\phi)]$
	fcc	$[0.4719 + 0.2058 f(\theta,\phi)]$ $[1.0742 + 0.4594 f(\theta,\phi)]$	$[0.6592 - 0.2569 f(\theta,\phi)]$ $[1.0742 + 0.4594 f(\theta,\phi)]$	$[0.8623 + 0.3363 f(\theta,\phi)]$ $[1.0742 + 0.4594 f(\theta,\phi)]$	$[0.9248 + 0.3832 f(\theta,\phi)]$ $[1.0742 + 0.4594 f(\theta,\phi)]$
$\langle a - p \rangle_{\theta,\phi}$ [see Sect. 8.2)	bcc	0.411	0.450	0.686	0.778
	fcc	0.441	0.600	0.785	0.854
$\frac{1}{T_{1\rho}}$ for $H_1^2 \gg H_{L\rho}^2$ in units of $\frac{\delta a_0^6}{H_1^2 \tau}$ [see (8.3)]	bcc	$[0.0911 + 0.0726 f(\theta,\phi)]$	$[0.1100 - 0.0656 f(\theta,\phi)]$	$[0.1676 + 0.1000 f(\theta,\phi)]$	$[0.1676 + 0.1440 f(\theta,\phi)]$
	fcc	$[0.4719 + 0.2058 f(\theta,\phi)]$	$[0.6592 - 0.2569 f(\theta,\phi)]$	$[0.8623 + 0.3363 f(\theta,\phi)]$	$[0.9248 + 0.3832 f(\theta,\phi)]$
$\frac{1}{T_{1\rho}}_{\theta,\phi}$ for $H_1^2 \gg H_{L\rho}^2$ in units of $\frac{\delta a_0^6}{H_1^2 \tau}$ (see Sec. VIII B)	bcc	0.1492	0.1625	0.2476	0.2828
	fcc	0.6365	0.8647	1.1313	1.2314

$(\tau \gg T_2^{\tau l})$. Then the "encounter" is reduced to a single jump. The only difference to the vacancy mechanism is that now spin i has no vacancy next to it. Thus we obtain in analogy to the derivation of Eq. (4.7),

$$1/T_c = 2(1 - p)/\tau ,\qquad (5.1)$$

with

$$p = \frac{1}{G}\sum_{i=1}^{G}\sum_{\vec{r}_m^0}F_{im}^{(0)}(\vec{r}_m^0)\,F_{im}^{(0)}(\vec{r}_m^0 - \vec{r}_i)\Big/\sum_{\vec{r}_m^0}|\,F_{im}^{(0)}(\vec{r}_m^0)\,|^2 . \qquad (5.2)$$

A more realistic random-walk model of diffusion than the one described in Refs. 8 and 4 would be to assume the nuclei to migrate on a sublattice only (so-called uncorrelated interstitial diffusion). In the SA region the relaxation rate for this mechanism was calculated by Ailion and Ho.[7] In the high-field region this mechanism has not yet been taken into account.

VI. EXTRAPOLATION TO THE HIGH-FIELD CASE

As mentioned earlier the SA theory is restricted to low fields, i.e., to the case $H_1 \le H_{L\rho}$. The reason is that in high fields $(H_1 \gg H_{L\rho})$ the splitting of the Zeeman levels becomes large compared to the local-field splitting, and a common spin temperature can no longer be established for the Zeeman and the total dipolar system.

This difficulty may be overcome by the use of perturbation theory for calculating high-field spin lattice relaxation in the roatating frame. Recently, the influence of correlated self-diffusion mechanisms on the high field $T_{1\rho}$ was taken into account by the present author.[4] For the low-temperature asymptote of the $T_{1\rho}$ minimum (i.e., for $\omega_1\tau \gg 1$), the following relationship was deduced for vacancy-induced self-diffusion:

$$\frac{1}{T_{1\rho}} = \frac{3}{8}\gamma^4\hbar^2 I(I+1)\,\frac{1}{\omega_1^2\tau}\sum_{\vec{r}_m^0}\Big(\,|\,F_{im}^{(0)}(\vec{r}_m^0)\,|^2 - F_{im}^{(0)}(\vec{r}_m^0)\sum_{\vec{r}_m^*}W_1(\vec{r}_m^0,\vec{r}_m^*)\,F_{im}^{(0)}(\vec{r}_m^0 + \vec{r}_m^*)\Big)\Big/[Z(0) + Z(\vec{r}_m^0)] . \qquad (6.1)$$

In Ref. 4 it was shown that (6.1) includes Torrey's theory of random-walk diffusion[8] as a special case, giving (for $\omega_1\tau \gg 1$)

$$\frac{1}{T_{1\rho}} = \frac{3}{8}\gamma^4\hbar^2 I(I+1)\,\frac{1}{\omega_1^2\tau}\sum_{\vec{r}_m^0}\Big(\,|\,F_{im}^{(0)}(\vec{r}_m^0)\,|^2 - F_{im}^{(0)}(\vec{r}_m^0)\frac{1}{G}\sum_{i=1}^{G}F_{im}^{(0)}(\vec{r}_m^0 - \vec{r}_i)\Big) , \qquad (6.2)$$

where $g(=1$ to $G)$ denotes the nearest-neighbor sites in the lattice.

In the rest of this section it will be shown that (6.1) and (6.2) may also be obtained from the results of Secs. III and V by means of a modification for high fields.

The total rotating-frame Hamiltonian $\mathcal{K}^0_{d_r}$ given by (3.1) may be regarded as a sum of two terms (see, e.g., Ref. 5)

$$\mathcal{K}^0_{d_r} = \mathcal{K}^{0,0}_{d_r} + \mathcal{K}^{0,n}_{d_r} , \tag{6.3}$$

where $\mathcal{K}^{0,0}_{d_r}$ denotes that part of $\mathcal{K}^0_{d_r}$ which commutes with the Zeeman Hamiltonian $\mathcal{K}_{z_r}$, while $\mathcal{K}^{0,n}_{d_r}$ denotes the noncommuting part. In high fields $(H_1 \gg H_{L\rho})$ the cross relaxation, by which the commuting Hamiltonians $\mathcal{K}^0_{d_r}$ and $\mathcal{K}_{z_r}$ are coupled together, becomes very slow. Within a time interval which is short compared to the cross-relaxation time T_{cr} sudden changes of $\mathcal{K}^0_{d_r}$ (resulting from nuclear or point-defect jumps) are communicated to $\mathcal{K}_{z_r}$ (and thus to the bulk magnetization) only via the noncommuting part $\mathcal{K}^{0,n}_{d_r}$, so that for $\tau \ll T_{cr}$ a common spin temperature may only be established between the noncommuting Hamiltonians $\mathcal{K}^{0,n}_{d_r}$ and $\mathcal{K}_{z_r}$. As shown by Ailion[5] this effect may be taken into account by replacing $H^2_{L\rho}$ by $\frac{3}{4} H^2_{L\rho}$ in Eq. (2.4).

Thus we obtain from (3.16) for vacancy-induced self-diffusion in the high-field region $(H_1 \gg H_{L\rho})$:

$$1/T_{1\rho} = \tfrac{3}{4} (H^2_{L\rho}/H^2_1) \, 2(a-p)/\tau . \tag{6.4}$$

Inserting $H^2_{L\rho}$ from Eq. (3.19) into (6.4), and making use of the relationship $\omega_1 = \gamma H_1$, we obtain

$$\frac{1}{T_{1\rho}} = \tfrac{3}{8} \gamma^4 \hbar^2 I(I+1) \frac{1}{\omega^2_1} \frac{a-p}{\tau} \sum_{\vec{r}^0_m} | F^{(0)}_{lm}(\vec{r}^0_m) |^2 . \tag{6.5}$$

Substituting a and p as given by Eqs. (3.17) and (3.18) into (6.5), we find Eq. (6.1), the result obtained from the application of perturbation theory.

Torrey's result (6.2) is derived similarly: as shown in Sec. V, for random-walk self-diffusion we have $a = 1$, and p is given by (5.1). Inserting these quantities into Eq. (6.5), we obtain (6.2).

VII. APPLICATION TO CUBIC CRYSTALS

Inserting a, p, and $H^2_{L\rho}$ from Eqs. (3.17)–(3.19) into (3.16), we obtain

$$\frac{1}{T_{1\rho}} = \frac{2}{\tau} \frac{\sum_{\vec{r}^0_m} \{[| F^{(0)}_{lm}(\vec{r}^0_m) |^2 - F^{(0)}_{lm}(\vec{r}^0_m) \sum_{\vec{r}^*_m} W_1(\vec{r}^0_m, \vec{r}^*_m) F^{(0)}_{lm}(\vec{r}^0_m + \vec{r}^*_m)]/[Z(0) + Z(\vec{r}^0_m)]\}}{H'^2_1 + \sum_{\vec{r}^0_m} | F^{(0)}_{lm}(\vec{r}^0_m) |^2} , \tag{7.1}$$

where the "reduced" field H'_1 is defined by

$$\tfrac{1}{4} \gamma^2 \hbar^2 I(I+1) a_0^{-6} H'^2_1 = H^2_1 . \tag{7.2}$$

According to the procedure described in Ref. 4 for the evaluation of lattice sums in cubic crystals, we may write for the lattice sums governing Eq. (7.1),

$$\sum_{\vec{r}^0_m} | F^{(0)}_{lm}(\vec{r}^0_m) |^2 = A_0 + B_0 f(\theta, \phi) , \tag{7.3}$$

$$\sum_{\vec{r}^0_m} \frac{| F^{(0)}_{lm}(\vec{r}^0_m) |^2}{Z(0) + Z(\vec{r}^0_m)} = A_1 + B_1 f(\theta, \phi) , \tag{7.4}$$

$$\sum_{\vec{r}^0_m} \frac{F^{(0)}_{lm}(\vec{r}^0_m) \sum_{\vec{r}^*_m} W_1(\vec{r}^0_m, \vec{r}^*_m) F^{(0)}_{lm}(\vec{r}^0_m + \vec{r}^*_m)}{Z(0) + Z(\vec{r}^0_m)}$$
$$= A_2 + B_2 \cdot f(\theta, \phi), \tag{7.5}$$

with

$$f(\theta, \phi) = (\sin 2\theta)^2 + \sin^4\theta \, (\sin 2\phi)^2 , \tag{7.6}$$

where the angles θ and ϕ characterize the crystallographic orientation of the field $\vec{H}_0$ (see Ref. 4).

Substituting (7.3) to (7.5) in (7.1), we get

$$\frac{1}{T_{1\rho}} = \frac{2}{\tau} \frac{A_1 - A_2 + f(\theta, \phi)(B_1 - B_2)}{H'^2_1 + A_0 + f(\theta, \phi) \cdot B_0} . \tag{7.7}$$

Equation (7.7) was deduced for vacancy-induced diffusion in the high-temperature region $(\tau_v \ll T_2^{r'} \ll \tau)$. Similar relationships are obtained for other diffusion mechanisms (see, e.g., Secs. IV and V and Ref. 7). Thus, we write for an arbitrary diffusion mechanism

$$\frac{1}{T_{1\rho}} = \frac{2}{\tau} \frac{A + f(\theta, \phi) B}{H'^2_1 + A_0 + f(\theta, \phi) B_0} . \tag{7.8}$$

In the high-field limit we obtain from the results of Sec. VI and from Eqs. (7.3)–(7.5):

$$\frac{1}{T_{1\rho}} = \frac{3}{2\tau} \frac{A + f(\theta, \phi) B}{H'^2_1} . \tag{7.9}$$

Inserting H'^2_1 given by (7.2) into (7.9), we have

$$\frac{1}{T_{1\rho}} = \frac{3}{8} \frac{\gamma^2 \hbar^2 I(I+1)}{a_0^6 H^2_1} [A + f(\theta, \phi) B] . \tag{7.10}$$

Numerical values of A, B, A_0, and B_0, calculated for the diffusion mechanisms treated in Secs. III–V, are listed in Sec. VIII.

TABLE II. Prefactors governing the orientation dependence of $T_{1\rho}$ [see Eqs. (7.3)–(7.7)] for vacancy diffusion in the high-temperature region (in units of a_0^{-6}; $2a_0$ = cube edge of a unit cell).

	A_0	B_0	A_1	B_1	A_2	B_2
bcc lattice	0.2324[a]	0.1622[a]	0.1291	0.0789	0.0380	0.0063
fcc lattice	1.0742[a]	0.4594[a]	0.5913	0.2354	0.1194	0.0296

[a]These values are in agreement with those published by Ailion and Ho (Ref. 7). It has to be noted that their values are presented in units of $16a_0^{-6}$.

VIII. NUMERICAL RESULTS

In Ref. 4, numerical values of $W_1(\vec{r}_m^0, \vec{r}_m^*)$ and $Z(\vec{r}_m^0)$ [see Eqs. (7.4) and (7.5)] were calculated by means of the computer simulation of an encounter (so-called Monte Carlo method). Thus, the relaxation rates (7.7) and (7.10) may be determined numerically.

A. Single crystals

In this section the orientation dependence of $T_{1\rho}$ given for a low field by (7.7) and for a high field by (7.10) is analyzed in some detail. Table II shows the numerical values of the prefactors A_i and B_i [see Eqs. (7.3)–(7.5) and Ref. 4].

For $H_1 = 0$ we obtain from Eqs. (3.16) and (7.8),

$$1/T_{1\rho} = 2(a - p)/\tau ,\qquad (8.1)$$

with

$$a - p = \frac{A + Bf(\theta, \phi)}{A_0 + B_0 f(\theta, \phi)} .\qquad (8.2)$$

Table I shows numerical values of A and B for the diffusion models described in this paper. In addition to the single-crystal low- and high-field cases [see Eqs. (8.1) and (7.10)] Table I includes numerical values for polycrystalline samples which were calculated according to the procedure described in Sec. VIII B.
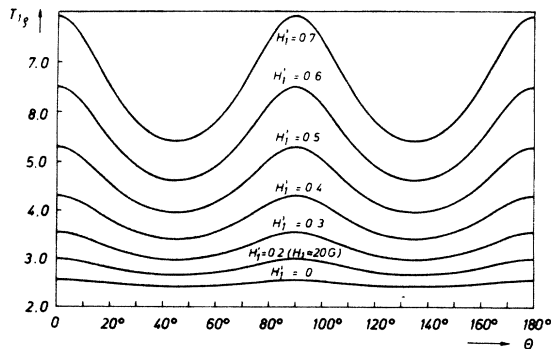
The variation of $(a - p)^{-1}$ [see Eq. (8.2)] for vacancy diffusion in the high-temperature region as a function of θ and ϕ is shown in Fig. 1. To demonstrate the strong increase of the orientation dependence of $T_{1\rho}$ with increasing field H_1, in Fig. 2 we have plotted $T_{1\rho}$ for different values of H_1'.

Another interesting effect on the orientation dependence of $T_{1\rho}$ is expected as the temperature is increased from the low-temperature region ($\tau_v \gg T_2^{rl}$) to the high-temperature region ($\tau_v \ll T_2^{rl} \ll \tau$). This effect is demonstrated in Fig. 3 for both zero field and for the high-field limit for $\phi = 0°$ and $\phi = 45°$ (see also Table I). As we see from Fig. 3 a variation of ϕ between 0° and 45° has different effects on the relaxation times in the two regions: while in the low-temperature region $T_{1\rho}$ should increase with increasing values of ϕ (for a given angle θ), the opposite is expected in the high-temperature region. This effect is particularly strong for values of θ close to 90°.

B. Polycrystalline samples

The relaxation behavior for polycrystalline samples is obtained from the single crystal results by

FIG. 2. Orientation dependence of $T_{1\rho}$ for $\phi = 0$ and different values of H_1' [in units of $\frac{1}{2}\tau$; see Eq. (7.8)]. H_1' is related to H_1 by Eq. (7.2) (for typical values of γ and a_0 we obtain $H_1^2 \approx 10^4 H_1'^2$).
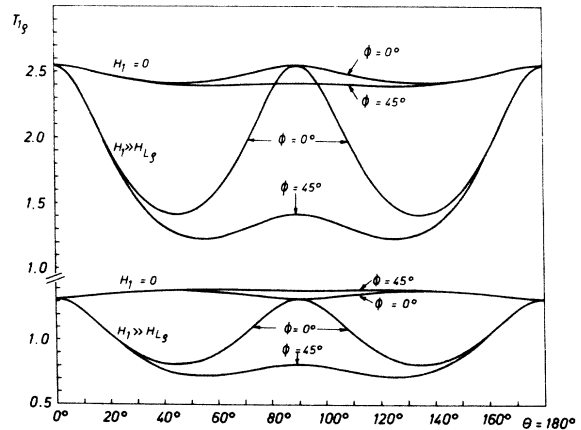

FIG. 3. Temperature dependence of the variation of $T_{1\rho}$ with the crystallographic orientation of the magnetic field for $H_1 = 0$ and $H_1 \gg H_{L\rho}$. Bottom: low-temperature region ($\tau_v \gg T_2^{rl}$). Top: high-temperature region ($\tau_v \ll T_2^{rl} \ll \tau$). (zero-field curves in units of $\frac{1}{2}\tau$; high-field curves in arbitrary units).

averaging the exponentially decaying magnetizations $M(\theta, \phi)$ in the individual crystallites over all orientations of θ and ϕ, i.e.,

$$\langle M(\theta, \phi) \rangle_{\theta, \phi} = \frac{M_0}{4\pi} \int_{\theta=0}^{\pi} \int_{\phi=0}^{2\pi} \exp\left(-\frac{t}{T_{1\rho}(\theta, \phi)}\right)$$

$$\times \sin\theta \, d\theta \, d\phi \ . \tag{8.3}$$

In general, Eq. (8.3) does not describe a simple exponential decay of $\langle M(\theta, \phi) \rangle_{\theta, \phi}$ [in Eq. (8.3) assumed to decay to zero]. It was shown in Ref. 4,

however, that the deviations from an exponential, characterized by the decay constant

$$\left\langle \frac{1}{T_{1\rho}(\theta, \phi)} \right\rangle_{\theta, \phi} = \frac{1}{4\pi} \int_{\theta=0}^{\pi} \int_{\phi=0}^{2\pi} \frac{1}{T_{1\rho}(\theta, \phi)} \sin\theta \, d\theta \, d\phi \ , \tag{8.4}$$

are very small for spin-lattice relaxation and may be neglected. Substituting Eq. (7.8) into (8.4), we obtain

$$\left\langle \frac{1}{T_{1\rho}} \right\rangle_{\theta, \phi} = \frac{2}{\tau} \frac{1}{4\pi} \int_{\theta=0}^{\pi} \int_{\phi=0}^{2\pi} \frac{[A + B f(\theta, \phi)] \sin\theta \, d\theta \, d\phi}{H_1'^2 + A_0 + B_0 f(\theta, \phi)} \ . \tag{8.5}$$

With $f(\theta, \phi)$ given by (7.6), we thus have

$$\left\langle \frac{1}{T_{1\rho}} \right\rangle_{\theta, \phi} = \frac{2}{\tau} \frac{1}{4\pi} \int_{\theta=0}^{\pi} \int_{\phi=0}^{2\pi} \frac{[A + B(\sin 2\theta)^2 + B\sin^4\theta(\sin 2\phi)^2] \sin\theta \, d\theta \, d\phi}{H_1'^2 + A_0 + B_0 \sin^2 2\theta + B_0 \sin^2 2\phi \sin^4 \theta} \ . \tag{8.6}$$

Integrating first with respect to ϕ, we get

$$\left\langle \frac{1}{T_{1\rho}} \right\rangle_{\theta, \phi} = \frac{2}{\tau} \left\{ \frac{B}{B_0} + \frac{1}{2} \left(\frac{A}{H_1'^2 + A_0} - \frac{B}{B_0} \right) \int_{\theta=0}^{\pi} \frac{\sin\theta \, d\theta}{\left[\left(1 + \frac{B_0}{H_1'^2 + A_0} \sin^2 2\theta \right) \left(1 + \frac{B_0}{H_1'^2 + A_0} (\sin^2 2\theta + \sin^4 \theta) \right) \right]^{1/2}} \right\} \tag{8.7}$$

Substituting $\cos\theta = x$ in Eq. (8.7) we obtain the following hyperelliptic integral for the relaxation rate:

$$\left\langle \frac{1}{T_{1\rho}} \right\rangle_{\theta, \phi} = \frac{2}{\tau} \left[\frac{B}{B_0} + \frac{1}{2} \left(\frac{A}{H_1'^2 + A_0} - \frac{B}{B_0} \right) \int_{-1}^{+1} \frac{dx}{[(1 + \epsilon) + (4\epsilon^2 + 6\epsilon) x^2 + (4\epsilon^2 - 7\epsilon) x^4 - 20\epsilon^2 x^6 + 12\epsilon^2 x^8]^{1/2}} \right], \tag{8.8}$$

where

$$\epsilon = B_0 / (H_1'^2 + A_0) \ . \tag{8.9}$$

So far we have not been able to solve the integral in Eq. (8.8) analytically and thus, in analogy to the results for single crystals [see (7.8)] to derive a linear dependence of $\langle T_{1\rho} \rangle_{\theta, \phi}$ on $H_1'^2$ (i.e., on H_1^2). However, from the numerical integration of (8.8) for different values of $H_1'^2$ it was found that $\langle T_{1\rho} \rangle_{\theta, \phi}$ is proportional to $H_1'^2$ even for very low fields $\{H_1'^2 = 10^{-6}$ to 10^{-4}, which typically corresponds to values of H_1^2 [see Eq. (7.2)] from about 10^{-2} G to 1 G$\}$. The values for polycrystalline samples listed in Table I were also obtained from the numerical integration of Eq. (8.8) (for $H_1'^2 = 0$ and $H_1'^2 \gg A_0$).

ACKNOWLEDGMENTS

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