PHYSICAL REVIEW B VOLUME 51, NUMBER 22 1 JUNE 1995-II

Spin dynamics in the amorphous antiferromagnet Si:P

M. J. R. Hoch and R. J. Hoch
*Department of Physics and Condensed Matter Physics Research Unit, University of the Witwatersrand,
Johannesburg, 2001, South Africa*

(Received 8 November 1994)

Heavily doped Si:P may be regarded as a disordered antiferromagnet. The spin dynamics have previously been investigated at low temperatures by means of ^{29}Si NMR relaxation-time measurements. Using detailed relaxation-time expressions, which take into account spin diffusion, the spectral functions extracted from the T_1 data, obtained as a function of applied field at different temperatures, are found to have the form $J(\omega)\sim\omega^{-\alpha}$, with $\alpha<1$ for $T\geq 1$ K and $\alpha\simeq 1$ for $T\ll 1$ K. The spectral function form at the higher temperatures is explained by means of a model, involving localized moments, linked to the Bhatt-Lee scaling theory for the spin susceptibility. The model requires adaptation to account for the very low temperature results.

I. INTRODUCTION

Following extensive studies of the magnetic properties of metal-insulator (MI) systems, in particular Si:P, it is generally accepted that the magnetic susceptibility behavior on the insulating side of the transition can be understood in terms of localized moment models such as the Bhatt-Lee[1] (BL) model, in which a number of the localized electron spins are "frozen" in a singlet pair state and do not contribute to χ at low temperatures. The total number of spins is replaced by a smaller effective number of spins, obtained using numerical calculations which involve renormalization procedures.

On the just-metallic side of the transition, the situation is more complicated and no complete theoretical explanation is available at present. A phenomenological two-fluid model, involving localized and delocalized spins, has, however, had some success in this region.[2,3]

The dynamics of MI systems have been probed on both the insulating[4,5] and just-metallic sides of the transition at low temperatures using nuclear magnetic resonance methods. Information on the spectral density, suggesting a $1/\omega$ frequency dependence, has been obtained. It is of interest to establish whether this dependence can be understood within the general framework of the BL localized moment model. The results of calculations which bear on this point are presented below and may prove useful for other systems classified as amorphous antiferromagnets.

II. THEORY: SPIN FLUCTUATIONS AND THE SPECTRAL DENSITY FUNCTION

Hoch and Holcomb[5] have suggested that localized electron-spin fluctuations are responsible for nuclear relaxation at low temperatures in Si:P and Si:(P,B). It is likely that below 4 K the spectral density of these spin fluctuations originates in the exchange coupling.

For very dilute paramagnetic systems, relaxation of the nuclear spins surrounding a paramagnetic ion is produced by a well-known mechanism.[6-8] Rorschach[8] has given the following general expression for the nuclear relaxation rate in such systems:

$$1/T_1=\frac{8\pi}{3}n\beta D f(\delta)\ , \tag{1}$$

where n is the concentration of impurities, D is the nuclear spin diffusion coefficient,

$$\beta=(C/D)^{1/4}\ , \tag{2}$$

with

$$C=\tfrac{2}{5}\gamma_S^2\gamma_I^2\hbar^2 S(S+1)J(\omega) \tag{3}$$

and

$$\delta=\beta^2/2b^2\ . \tag{4}$$

The quantity b, the diffusion barrier radius, plays an important role in analyzing results using Eq. (1).

The function,

$$f(\delta)=I_{3/4}(\delta)/I_{-3/4}(\delta)\ , \tag{5}$$

where the I_m are modified Bessel functions, takes the following limiting values:

$$\delta\gg 1:\ \ f(\delta)\simeq 1\ \ \text{(diffusion-limited regime)}\ ,$$

$$\delta\ll 1:\ \ f(\delta)\simeq 2^{-2/3}\delta^{3/2}\ \ \text{(rapid diffusion regime)}\ .$$

Correspondingly, we obtain

$$\delta\gg 1:\ \ 1/T_1=8\pi/3nC^{1/4}D^{3/4}\ , \tag{6a}$$

$$\delta\ll 1:\ \ 1/T_1=4\pi/3nb^{-3}C\ . \tag{6b}$$

The spin diffusion coefficient D can be obtained from the relationship $D=a^2(M_2)^{1/2}/30$, with a the internuclear spacing and M_2 the nuclear dipolar second moment.

The spectral function $J(\omega)$ in Eq. (3) can, in principle, be studied by making nuclear relaxation-time measurements as a function of frequency. From a theoretical point of view, $J(\omega)$ may be obtained from the spin correlation function $G(t)$. For very dilute systems, in which

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interactions between the paramagnetic ions are negligibly weak, an exponential form is usually chosen for $G(t)$ and the Fourier transform gives the Debye form for $J(\omega)$. $G(t)$ is related to the imaginary part of the dynamical magnetic susceptibility χ'' by the fluctuation-dissipation theorem.

For systems containing interacting paramagnetic ions, it is necessary to allow for electron spin fluctuations due to spin-spin interactions within the exchange-coupled reservoir. The total Hamiltonian for the system may be written as

$$\mathcal{H}_0=\mathcal{H}_I^Z+\mathcal{H}_I^D+\mathcal{H}_S^Z+\mathcal{H}_S^E+\mathcal{H}_{IS}^P+\mathcal{H}_{IS}^{\mathrm{hf}} , \tag{7}$$

where $\mathcal{H}_I^Z$ and $\mathcal{H}_S^Z$ are the I-spin and S-spin Zeeman Hamiltonians, respectively, $\mathcal{H}_I^D$ is the nuclear dipolar Hamiltonian, and $\mathcal{H}_S^E=\sum_{i<j}J_{ij}S_i$. S_j is the S-spin exchange Hamiltonian, and

$$\mathcal{H}_{IS}^P=\sum_{i,\mu}B_{i\mu}S_{iz}I_\mu^\pm \tag{8}$$

is the nonsecular term in the dipolar coupling between I and S and is responsible for I-spin relaxation.

In Eq. (8),

$$B_{i\mu}=\tfrac{3}{2}\gamma_I\gamma_S\hbar\frac{\sin\theta_{i\mu}\cos\theta_{i\mu}}{r_{i\mu}^3} , \tag{9}$$

where $r_{i\mu}$ is the vector joining the ith S spin to the μth I spin and $\theta_{i\mu}$ is the angle this vector makes with the applied magnetic field. We neglect the small dipolar Hamiltonian $\mathcal{H}_I^D$ in comparison to $\mathcal{H}_I^Z$. $\mathcal{H}_I^D$ is important, however, in maintaining a spin temperature in the I spin system through spin diffusion. We also neglect the hyperfine coupling $\mathcal{H}_{IS}^{\mathrm{hf}}$ for the nuclear spins of interest, which are at some distance from the S spin. This interaction is discussed in detail later in connection with the quantity b in Eq. (4).

With the above assumptions, using the master equation for the density matrix,[6] we obtain the following expression for the transition rate for I spins:

$$W=\frac{\mathrm{Tr}\{(\mathcal{H}_{IS}^P)^2\}}{\mathrm{Tr}\{I_Z^2\}}\int_0^\infty G(t)\cos(\omega_I t)dt , \tag{10}$$

where $\omega_I=\gamma_I B_0$ and

$$G(t)=\frac{\mathrm{Tr}\{\mathcal{H}_{IS}^P\exp(-i\mathcal{H}_S^E t)\mathcal{H}_{IS}^P\exp(i\mathcal{H}_S^E t)\}}{\mathrm{Tr}\{(\mathcal{H}_{IS}^P)^2\}} . \tag{11}$$

Evaluating the traces gives, for the transition rate,

$$W=\frac{2}{3}\sum B_{i\mu}^2(r_{i\mu},\theta_{i\mu})S(S+1)J(\omega_I) , \tag{12}$$

where

$$J(\omega_I)=\int_0^\infty G(t)\cos\omega_I t\, dt \tag{13}$$

is the cosine transform of the correlation function $G(t)$.

Doped semiconductors may be regarded as dilute amorphous anitferromagnets. The form of $G(t)$ for these systems is not known. If both spin-lattice and spin-spin interactions are important, assuming that the competing mechanisms are independent, we can write the correlation function as a product,

$$G(t)=G_{SS}(t)G_{SL}(t) , \tag{14}$$

where $G_{SS}(t)$ is the spin-spin correlation function and $G_{SL}(t)$ is the spin-lattice correlation function. We shall focus on $G_{SS}(t)$ and assume that the temperature is sufficiently low that $G_{SL}(t)$ makes an unimportant contribution to $G(t)$. Generalization to the case where this is not so is straightforward.

Goldman, Cox, and Bouffard[9] and Melikeya[10] have suggested that the spectral function for F center spins in LiF may be represented by a truncated Lorentzian curve. This corresponds to a modified exponential correlation function. In cross relaxation between rare and abundant nuclear spin species in $\mathrm{CaF_2}$, McArthur, Hahn, and Walstedt[11] have found that a Lorentzian correlation function provides a good description of the dipolar fluctuation spectrum for the ^{19}F spins. Lorentzian correlation functions have subsequently been shown to be important in cross relaxation in a number of other systems.[12] Demco, Tegenfeldt, and Waugh,[13] in work on cross relaxation in systems containing two nuclear spin species, have provided further theoretical and experimental support for the Lorentzian form for the rotating-frame adiabatic demagnetization case. They obtain a Gaussian form for the spin-locked case. For exponential, Gaussian, and Lorentzian correlation functions, we find

$$\left.\frac{d^2G}{dt^2}\right|_{t=0}=\frac{a}{\tau^2} , \tag{15}$$

where a is a constant of order unity.

Using Eq. (11) together with the result in Eq. (15) gives

$$\frac{a}{\tau^2}=-\frac{\mathrm{Tr}\{\mathcal{H}_{IS}^P,\mathcal{H}_E\}^2}{\mathrm{Tr}\{\mathcal{H}_{IS}^P\}} . \tag{16}$$

Evaluating the traces yields

$$\frac{a}{\tau^2}=\tfrac{2}{3}I(I+1)\frac{\sum_{i<j}J_{ij}^2(B_i-B_j)^2}{\sum B_i^2} . \tag{17}$$

This expression may be interpreted as follows: a pair of electrons i and j will spin exchange at a rate given by $J_{ij}/\hbar$. The factor (B_i-B_j) gives a measure of the importance of spins i and j in producing relaxation effects at the lattice site μ, where a nucleus is situated. For a symmetric situation, with $B_i=B_j$, no relaxation is produced by the exchange process. Such a situation is highly unlikely for most of the nuclear spins in the system. It is probable that, for a given nucleus μ, a single S spin i will dominate, with $r_{i\mu}<r_{j\mu}$, for all $j\neq i$. In this case, Eq. (17) may be simplified to give, for $I=\frac{1}{2}$,

$$\frac{a}{\tau^2}\simeq\frac{J^2}{2\hbar^2} . \tag{18}$$

If we integrate over a volume $V=1/n_s$, where n_s is the number of S spins per unit volume, and put $1/T_1=2W$, we obtain Eq. (6b), corresponding to the direct relaxation or diffusion-limited regime. Equation (6a) can be obtained for the rapid diffusion case. $J(\omega)$ is given by Eq. (13), with the correlation time obtained from Eq. (18).

For the exchange-coupled disordered system of electron spins, there is a distribution of J values for spin pairs. In the BL model for magnetic susceptibilities, the form $P(J)\propto J^{-\alpha}$ is assumed to hold over a range of J values sufficient for the calculations. The exponent α takes values in the range 0.8–0.6, decreasing as n_s tends toward n_c. Tightly coupled spin pairs, with $J>k_BT/\hbar$, are regarded as "frozen" in the singlet state. Correspondingly, their contribution to the spectral function will be limited to rather high frequencies ($\sim 10^{10}$ Hz) for $T\sim 1$ K.

Experiments have probed the low-frequency part of the spectrum where, in general, $\omega \ll k_BT/\hbar$. Figure 1 schematically illustrates the form of the J distribution and the various frequencies of interest.

In order to proceed, we assume an exponential form for the pair correlation function $G(t)$. This has the advantage of simplicity, and calculations have shown that the ensemble average spectral density for all pairs is insensitive to the form chosen for $G(t)$ when integration is carried out over a distribution of τ values. If we convert the J distribution into a correlation time distribution using $\tau\sim\hbar/J$ and $P(J)\sim J^{-\alpha}$, as mentioned above, in connection with the BL model, we obtain

$$P(\tau)d\tau \simeq \frac{A}{\tau^{2-\alpha}}d\tau \tag{19}$$

for $\tau>\tau_{\min}\sim\hbar/k_BT$. A is a constant. Integrating over the τ distribution leads to the following expression for the ensemble average spectral function:

$$J(\omega)=\int_{\hbar/k_BT}^{\infty}\left[\frac{1}{(1+\omega_I^2\tau^2)\tau^{1-\alpha}}\right]d\tau \, . \tag{20}$$

For $\alpha=1$, we obtain

$$J(\omega)=\pi/2\omega-\hbar/k_BT \, . \tag{21}$$

This agrees with the result, given previously,[14,15] which is based on the *adhoc* assumption $P(\tau)\propto 1/\tau$. For $\alpha\neq 1$, the integral in Eq. (20) must be evaluated numerically. This has been done for a number of values of α. The results are presented below.

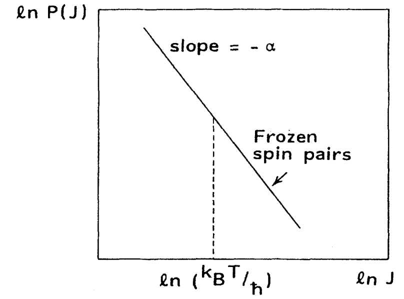

FIG. 1. Schematic illustration of the form of the J distribution $P(J)\sim J^{-\alpha}$. The cutoff value of $J=k_BT/\hbar$ separates "frozen" spin pairs and "unfrozen" pairs.

$J(\omega)$ may be deduced from the NMR relaxation rate measurements using Eq. (1), with D calculated using the expression quoted in Sec. II. The parameter that gives rise to the greatest uncertainty in determining $J(\omega)$ is b, which is a function of the applied magnetic field B. b may be estimated using the relationship

$$B(r)_{r=b}\simeq 2\pi/\gamma T_2\sim\Delta B_n \, , \tag{22}$$

where ΔB_n is the natural (dipolar) nuclear linewidth. Abragam and Goldman[12] have pointed out that a better estimate of b may be obtained using

$$a\frac{\partial}{\partial r}[B(r)]_{r=b}\simeq\Delta B_n \, , \tag{23}$$

where a is the nuclear spin spacing. The dipolar ($B_{\rm dip}$) and hyperfine ($B_{\rm hf}$) fields may be estimated using the following expressions:

$$B_{\rm dip}(r)\simeq\frac{\langle\mu_e\rangle}{r^3} \tag{24a}$$

and

$$B_{\rm hf}(r)\simeq\frac{8\pi}{3}\langle\mu_e\rangle|\psi_0|^2e^{-2r/b^*} \, , \tag{24b}$$

with

$$\langle\mu_e\rangle\simeq\mu_e\tanh\left[\frac{\mu_eB}{k_BT}\right] \, .$$

The impurity wave function parameters are $|\psi_0|^2=4.2\times10^{22}$ cm^{-3} and $b^*=15$ Å for Si:P. We obtain

$$b_{\rm dip}=\left[\frac{\gamma_S\gamma_I\hbar T_2\mu_eB}{2k_BT}\right]^{1/3}$$

$$\simeq 2.4\times10^{-8}[B/T]^{1/4}\ \text{cm} \tag{25}$$

and

$$b_{\rm hf}\simeq 7.5\times10^{-8}\ln[7.2\times10^3\tanh(6.72\times10^{-5}B/T)] \, . \tag{26}$$

At $T=1.3$ K, $b_{\rm hf}>b_{\rm dip}$ for all B values of interest.

It should be noted that, in the diffusion-limited region ($\delta\gg1$) for which Eq. (6b) applies,

$$J(\omega)\propto\left[\frac{3}{8\pi}nD^{3/4}\left[\frac{1}{T_1}\right]\right]^4 . \tag{27}$$

In the rapid diffusion regime ($\delta\ll1$),

$$J(\omega)\propto\frac{3b^3}{4\pi n}\left[\frac{1}{T_1}\right] . \tag{28}$$

For both expressions, n should be replaced by $n_{\rm eff}$, as discussed by Hoch and Holcomb.[5] This has the effect of scaling the spectral function, but does not change the form of the frequency dependence. In the crossover region $\delta\sim1$, no simple expression for $J(\omega)$ can be obtained and the full expression of Eq. (1) must be used to obtain $J(\omega)$ from the measured T_1 values. The modified Bessel functions are obtained from standard tables.

III. DISCUSSION

Figure 2 shows a plot of the high-field spin-lattice relaxation rate at 1.6 K (Ref. 14) as a function of concentration for $0.1 < n/n_c < 100$. Plotted on the same figure is the low-field magnetic susceptibility at 1.1 K.[15] The similarity in the shape of the curves is striking. This suggests a relationship between χ and $1/T_1$. Using arguments based on the fluctuation-dissipation theorem, the spectral function may be written as

$$J(\omega) \propto \chi''(\omega) \propto \chi_s f(\omega) ,$$

where $\chi''(\omega)$ is the imaginary part of the magnetic susceptibility, χ_s is the static susceptibility, and $f(\omega)$ is a shape function. From Eq. (6b), we note that $1/T_1 \propto J(\omega)$, provided $\delta \gg 1$, which is appropriate in the high-field case. Figure 2 therefore provides support for the present analysis of the relaxation-time data, based on the BL model ideas, which have been developed to account for the magnetic susceptibility data for $n/n_c \lesssim 1$. A quantitative analysis of the relationship between $1/T_1$ and χ_s on the insulating and metallic sides of the transition has not been attempted.

It is interesting to note that antiferromagnetic spin correlations are believed to be important in nuclear relaxation in the high-T_c cuprate superconductors.[16–18] For these systems, the spin-lattice relaxation rate may be expressed in terms of the dynamic susceptibility $\chi''(q,\omega)$.

$J(\omega)$ has been extracted from the available relaxation-time measurements, made as a function of frequency, using Eqs. (27) and (28) and the expressions quoted for b and D. The greatest uncertainty in this procedure is associated with the magnitude of b. Figure 3 shows a plot of b versus B for 1.3 K and 13.5 mK based on Eqs. (25) and (26). Both b_{dip} and b_{hf} are plotted, and it can be seen that b_{hf} is of dominant importance at both temperatures. At 13.5 mK, b_{hf} reaches a plateau value of 6.7 nm at fields of a few hundred gauss. The 1.3 K behavior for b_{hf} shows a gradual increase to around 6 nm at 1 T. These values are comparatively large, being close to the nearest-neighbor mean separation $\langle r_{\text{NN}} \rangle$ (6.5 nm) of impurity atoms for $n = n_c$. It is therefore possible that the procedure for obtaining $J(\omega)$ from the relaxation-rate data will become unreliable at the higher field values where $b_{\text{hf}} \simeq \langle r_{\text{NN}} \rangle$.

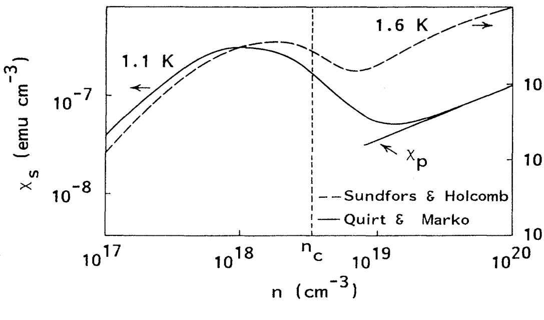

FIG. 2. Measured Si:P spin susceptibility χ_s at 1.1 K (Ref. 15) vs n_s plotted together with the measured relaxation rate for ^{29}Si at 1.6 K (Ref. 16).

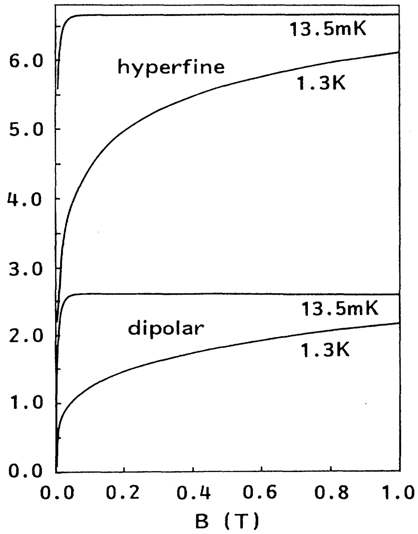

FIG. 3. Diffusion barrier radius predictions at 1.3 K and 13.5 mK corresponding to dipolar and hyperfine couplings between S and I spins.

Figure 4(a) shows $J(\omega)$, as a function of frequency, obtained from the ^{29}Si relaxation-time measurements[5,19] at 1.3 K for Si:P samples, with the n/n_c values shown in the caption. The n_{eff} values[5] used are consistent with the BL model. Figure 4(b) shows similar plots for data obtained at 13.5 mK for two samples.[4]

While caution must be exercised in interpreting the spectral function curves because of uncertainties in the quantity b, particularly at the highest fields used, some general conclusions can be stated. At 1.3 K, the spectral function appears to have the form $J(\omega) \propto \omega^{-\alpha}$, with $\alpha \sim 0.7$, for the n/n_c values shown, over much of the range of ω which has been explored. Departures from this form at the high- and low-frequency ends are probably connected to the uncertainties in b mentioned above. At this temperature it may be concluded that the dynamics of the system are consistent with the ideas of Sec. II, based on the BL model and the J distribution $P(J) \sim J^{-\alpha}$.

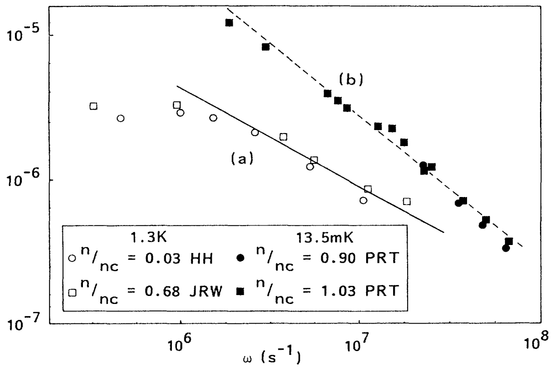

FIG. 4. Spectral functions $f(\omega)$ obtained from ^{29}Si relaxation data at (a) 1.3 K [HH (Ref. 5), JRW (Ref. 19)] and (b) 13.5 mK [PRT (Ref. 4)].

At 13.5 mK the spectral curve appears to have a value of α close to unity, so that $J(\omega) \propto \omega^{-1}$. It has previously been pointed out[5] that at 13.5 mK the electron-spin system is highly polarized. The relaxation mechanism may involve electron dynamics quite different from the spin exchange flip-flop processes which are important at higher temperatures. Comparison of Figs. 4(a) and 4(b) suggests that this may be so.

Relaxation processes in many amorphous systems have been found to be governed by a correlation function with a stretched exponential form[20] $G(t)=e^{-(t/\tau_c)^\beta}$. We have generated the corresponding spectral functions by means of Fourier transformation, but have not been able to fit the spectral curves in a satisfactory way for any choice of the parameter β.

It appears that at $T \sim 1$ K the spectral function describing the spin dynamics of the exchange-coupled reservoir is consistent with the ideas of Sec. III, based on the BL model and the distribution of J values. In particular, "frozen" spin pairs do not contribute to the spectral function at the frequencies used in the NMR experiments. Alternatively stated, it is the low-q modes which play a dominant role in relaxation. At temperatures much lower than 1 K, it is likely that the theory given in Sec. III cannot be applied and that the relaxation processes require a more elaborate description than is presented there. A probable reason for this is that the electron spins become highly polarized in the millikelvin temperature region.

IV. CONCLUSION

The spectral functions $J(\omega)$, describing exchange reservoir dynamics in the amorphous antiferromagnet Si:P system close to the MI transition, have been extracted from nuclear relaxation data obtained at two temperatures. Making use of Bhatt-Lee model ideas and integrating over a distribution of correlation times lead to the form $J(\omega) \propto \omega^{-\alpha}$, with $\alpha < 1$, which fits the experimentally derived curves quite well at 1.3 K. The strongly coupled "frozen" spin pairs do not contribute to the spectral function at frequencies probed in the nuclear relaxation experiments. The less strongly coupled spins play a dominant role in relaxation.

At temperatures much below 1 K, the $J(\omega)$ curves obtained from the relaxation data appear to have the form $J(\omega) \propto \omega^{-1}$. It seems likely that, for the strongly polarized spin systems, the dynamics are more complicated than at higher temperatures. Further work is required to determine the nature of the relaxation processes in the millikelvin range.

ACKNOWLEDGMENTS

We should like to express our thanks to Professor D. F. Holcomb of Cornell University for his helpful comments on the manuscript. Financial support from the Foundation for Research Development is gratefully acknowledged.

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