

# Theory of optical orientation in *n*-type semiconductors

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Time-resolved measurements of magnetization in *n*-GaAs have revealed a rich array of spin decoherence processes, and have shown that fairly long lifetimes ( $\sim 100$  ns) can be achieved under certain circumstances. In time-resolved Faraday rotation and time-resolved Kerr rotation the evolution of the magnetization can be followed as a function of temperature, applied field, doping level, and excitation level. We present a theory for the spin relaxation in *n*-GaAs based on a set of rate equations for two interacting thermalized subsystems of spins: localized states on donor sites and itinerant states in the conduction band. The conduction-band spins relax by scattering from defects or phonons through the D'yakonov-Perel' mechanism, while the localized spins relax by interacting with phonons (when in an applied field) or through the Dzyaloshinskii-Moriya interaction. In this model, numerous features of the data, including puzzling temperature and doping dependencies of the relaxation time, find an explanation.

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Spin coherence in semiconductors is attracting renewed attention due to the prospects of spintronics—information storage and processing using spin rather than charge degrees of freedom, and by the idea that spins in semiconductors could serve as qubits for quantum computers.<sup>1,2</sup> For qubit applications, the spin degrees of freedom must be coherent, which necessitates a detailed understanding of the processes that limit spin lifetimes. Time-resolved measurements on *n*-type systems have revealed an array of decoherence processes, and have shown that fairly long lifetimes (greater than 100 ns) can be achieved in *n*-GaAs.<sup>3,4</sup> In time-resolved Faraday rotation and time-resolved Kerr rotation the evolution of the magnetization can be followed as a function of temperature, applied field, doping level, and the intensity and duration of the pump pulse. Results on different materials (GaAs, GaN, ZnSe) are similar, pointing to universality in the phenomena. Awschalom and Samarth<sup>5</sup> have reviewed the experimental situation.

Our theory provides a systematic framework for investigating the wide range of parameters studied in optical orientation experiments on *n*-type semiconductors. Previous theories have concentrated on either higher temperatures,<sup>3</sup> or on very low temperatures and very low magnetic fields.<sup>4</sup> We account for certain puzzling experimental observations by having two distinct types of spin states: localized donor states and itinerant conduction-band states, with characteristic spin-relaxation rates  $1/\tau_l$  and  $1/\tau_c$ , respectively. In addition, a fast cross-relaxation rate,  $1/\tau_{cr}$ , between the localized and itinerant spins is a crucial feature of these systems, leading to the largest relaxation rate, either  $1/\tau_c$  or  $1/\tau_l$ , usually tending to dominate the spin dynamics for the whole system. Examples of this behavior are shown in Figs. 1 and 2 below. In this paper we focus on presenting the theory and applying it to *n*-GaAs, reserving a more extensive comparison to experiments for a later publication.

The cross relaxation between localized and itinerant spins occurs by the usual exchange interaction Hamiltonian

$$H_{l-c} = \frac{J}{V} \sum_{i,k} \vec{s}_i \cdot \vec{s}_k,$$

where the sum runs over impurity spins  $i$  and conduction-band states  $k$  and  $V$  is the volume of the system. This Hamiltonian conserves total spin and cannot, by itself, relax the magnetization. However, it can transfer spin from localized to itinerant states.  $J$  may be estimated as  $J \sim -e^2 a_B^2$ . Here  $a_B = 10.4$  nm is the effective Bohr radius for an impurity. If virtual excitations to the upper-Hubbard band are important, this estimate could be reduced and  $J$  could even change sign. The sign is actually not important for our purposes, since all experimental temperatures are well above the Kondo temperature. For *n*-GaAs an order-of-magnitude estimate for the

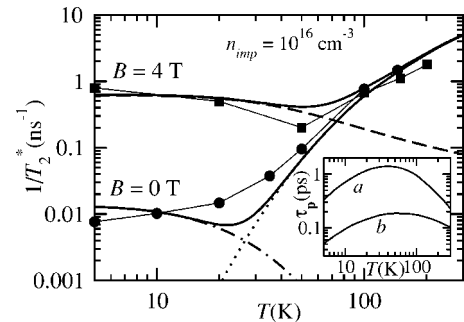


FIG. 1. Plot of  $1/T_2^*$  vs temperature. The data are from Ref. 3, with solid dots for  $B=0$  T and solid squares for  $B=4$  T. The lines connecting the data points are guides for the eye. The curves are a least-squares fit of Eq. (5) to the data. Dashed-dotted curve:  $(n_l/n_{imp})(1/\tau_{DM})$ ; dashed curve:  $(n_l/n_{imp})(1/\tau_{s-ph})$  for  $B=4$  T; dotted curve:  $(n_c/n_{imp})(1/\tau_c)$ ; solid curves: total  $1/T_2^*$ . For  $B=0$  T,  $1/\tau_{s-ph}=0$ . Inset:  $\tau_p$  vs temperature for (a)  $n_{imp}=10^{16}$   $\text{cm}^{-3}$  and (b)  $n_{imp}=10^{18}$   $\text{cm}^{-3}$ .

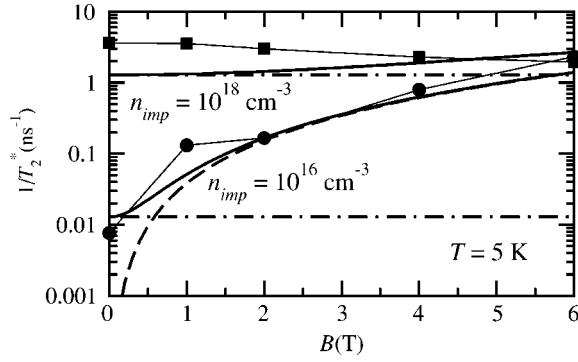


FIG. 2. Plot of  $1/T_2^*$  vs applied magnetic field. The data are from Ref. 3, with solid dots for  $n_{imp}=10^{16}$  cm $^{-3}$  and solid squares for  $n_{imp}=10^{18}$  cm $^{-3}$ . The lines connecting the data points are guides for the eye. The curves are a least-squares fit of Eq. (5) to the data. Dashed-dotted curves:  $(n_l/n_{imp})(1/\tau_{DM})$  for the two densities; dashed curve:  $(n_l/n_{imp})(1/\tau_{s-ph})$ ; solid curves: total  $1/T_2^*$ . For  $T=5$  K,  $(n_c/n_{imp})(1/\tau_c)\ll 0.001$  ns $^{-1}$ .

cross-relaxation rate is  $1/\tau_{cr}\sim(1\text{ ps}^{-1})(n_{imp}/n_0)$ , where  $n_0=10^{18}$  cm $^{-3}$  is a fiducial density.

The spin orientation is created by a circularly polarized optical pump pulse about 100-fs long tuned near the band gap, creating particle-hole pairs. The valence-band holes depolarize quickly and fast recombination (on a time scale of 50–100 ps) leaves the conduction-band and localized donor state system with a net spin polarization along the propagation direction of the beam ( $z$  direction). The time evolution of this polarization is tracked by applying a transverse magnetic field in the  $x$  direction (Voigt geometry). The resulting precession about the  $x$  axis and concomitant decay are measured optically, with  $1/T_2^*$  the relaxation rate of the macroscopic transverse magnetization.

Our theory may be formalized by writing modified Bloch equations for the magnetization for times after recombination ( $t>100$  ps after the end of the pump pulse). The holes have recombined and spin-conserving processes have thermalized the system, subject to the constraint that the magnetization retains the polarization produced by the initial excitation process. There are then two thermalized subsystems of electrons at ambient temperature with relative occupations determined by standard thermodynamic methods. The localization of conduction-band electrons onto impurity sites takes place at a temperature scale  $T_{imp}$  determined by  $n_{imp}$ . For  $n_{imp}=10^{16}$  cm $^{-3}$  we have  $T_{imp}\approx 50$  K. Denote the localized and conduction-band densities by  $n_l$  and  $n_c$ , with  $n_{imp}=n_l+n_c$ . In the experiments,  $N_{ex}$ , the density of electrons excited by the pump pulse, is small,  $N_{ex}\ll n_{imp}$ , except possibly for nominally insulating samples, which we discuss briefly below.

We work in the frame which rotates about the  $x$  axis at a rate  $g^*\mu_B B/\hbar$ , and is along the  $z$  axis at time  $t=0$ . In this frame, the dynamics are governed by

$$\frac{dn_{c+}}{dt} = -\frac{n_{c+}}{\gamma_{cr}}n_{l-} + \frac{n_{c-}}{\gamma_{cr}}n_{l+} - \frac{1}{2\tau_c}n_{c+} + \frac{1}{2\tau_c}n_{c-}, \quad (1)$$

$$\frac{dn_{c-}}{dt} = -\frac{n_{c-}}{\gamma_{cr}}n_{l+} + \frac{n_{c+}}{\gamma_{cr}}n_{l-} - \frac{1}{2\tau_c}n_{c-} + \frac{1}{2\tau_c}n_{c+}, \quad (2)$$

$$\frac{dn_{l+}}{dt} = -\frac{n_{l+}}{\gamma_{cr}}n_{c-} + \frac{n_{l-}}{\gamma_{cr}}n_{c+} - \frac{1}{2\tau_l}n_{l+} + \frac{1}{2\tau_l}n_{l-}, \quad (3)$$

$$\frac{dn_{l-}}{dt} = -\frac{n_{l-}}{\gamma_{cr}}n_{c+} + \frac{n_{l+}}{\gamma_{cr}}n_{c-} - \frac{1}{2\tau_l}n_{l-} + \frac{1}{2\tau_l}n_{l+}, \quad (4)$$

where  $+$  and  $-$  denote up and down spins in the rotating frame and  $\gamma_{cr}=n_0\tau_{cr}$ . By rewriting Eqs. (1)–(4) in terms of the total densities,  $n_l=n_{l+}+n_{l-}$  and  $n_c=n_{c+}+n_{c-}$ , and the magnetization densities,  $m_l=n_{l+}-n_{l-}$  and  $m_c=n_{c+}-n_{c-}$ , we find that the total densities are time independent,  $dn_l/dt=0$ , and  $dn_c/dt=0$ , and the magnetization densities are determined by

$$\frac{dm_c}{dt} = -\left(\frac{1}{\tau_c} + \frac{n_l}{\gamma_{cr}}\right)m_c + \frac{n_c}{\gamma_{cr}}m_l \quad (5)$$

$$\frac{dm_l}{dt} = \frac{n_l}{\gamma_{cr}}m_c - \left(\frac{1}{\tau_l} + \frac{n_c}{\gamma_{cr}}\right)m_l. \quad (6)$$

In general, the time dependence of the total magnetization  $m(t)=m_c(t)+m_l(t)$  is a sum of two exponentials,  $\exp(-\Gamma_+t)$  and  $\exp(-\Gamma_-t)$  (behavior observed in experiments on  $n$ -GaN)<sup>6</sup> with eigenvalues

$$\Gamma_{\pm} = \frac{1}{2}\left(\frac{1}{\tau_c} + \frac{1}{\tau_l} + \frac{n_{imp}}{\gamma_{cr}} \pm S\right), \quad (7)$$

where  $S$  is given by

$$S = \sqrt{\left(\frac{1}{\tau_l} - \frac{1}{\tau_c} + \frac{n_c - n_l}{\gamma_{cr}^2}\right)^2 + \frac{4n_l n_c}{\gamma_{cr}^2}}. \quad (8)$$

For  $n$ -GaAs, we are in the regime  $1/\tau_{cr}\gg 1/\tau_c, 1/\tau_l$ , where the eigenvalues give two very different relaxation rates: a very rapid relaxation given by  $\Gamma_+\approx n_{imp}/(n_0\tau_{cr})$ , with a timescale on the order of picoseconds, and a slower relaxation given by

$$\Gamma_- = \frac{1}{T_2^*} \approx \frac{n_l/n_{imp}}{\tau_l} + \frac{n_c/n_{imp}}{\tau_c}, \quad (9)$$

with a timescale on the order of tens of nanoseconds. Given the expressions for  $1/\tau_l$  and  $1/\tau_c$ , Eq. (9) gives the calculated total relaxation rate from our theory. This is the appropriate quantity to compare to the single exponential time dependence observed in experiments.<sup>3,4</sup>

There are various processes that can relax the nonequilibrium magnetization produced in optical orientation experiments. The conduction-band processes have been well studied,<sup>7</sup> while the relaxation mechanisms for localized electrons are less well understood.

**D'yakonov-Perel' Mechanism.** Conduction-band electron spins in  $n$ -GaAs relax primarily by the D'yakonov-Perel' (DP) mechanism,<sup>8</sup> due to lack of inversion symmetry in III-V systems. The lack of inversion symmetry, together with spin-orbit coupling, gives an effective  $\vec{k}$ -dependent magnetic field, causing the spin of an itinerant electron to precess about an axis related to  $\vec{k}$ . The precession frequency for an electron at wavenumber  $\vec{k}$  is  $\Omega_{DP}(\vec{k})$ , and the DP relaxation comes from switching the precession axis by scat-

tering from one  $\vec{k}$  vector to another. Assuming  $s$ -wave scattering, one obtains  $1/\tau_{DP}(\vec{k})=2\Omega_{DP}^2(\vec{k})\tau_p(\vec{k})/3$ , where  $\tau_p(\vec{k})$  is the momentum relaxation time. Averaging this expression over the Boltzmann distribution, and using the results of Fishman and Lampel for the momentum average,<sup>9</sup> we find a spin relaxation rate  $1/\tau_{DP}=\alpha_{DP}T^3\tau_p$ , with  $\alpha_{DP}(\text{th})=9.0 \times 10^{-10} \text{ K}^{-3}\text{ps}^{-2}$ . Here  $\tau_p$  is the average momentum relaxation time which has a complicated temperature and doping dependence (shown in Fig. 1) best taken from mobility data,  $\mu_e=e\tau_p/m^*$ .<sup>10</sup>

**Elliot-Yafet Mechanism.** Conduction-band electron spins can also relax via the Elliot-Yafet (EY)<sup>11</sup> mechanism, due to ordinary impurity scattering from state  $\vec{k}$  to state  $\vec{k}'$ . With spin-orbit coupling the initial and final eigenstates are not eigenstates of  $S_z$ , the spin projection operator, so this process relaxes the spin. One finds  $1/\tau_{EY}=\alpha_{EY}T^2/\tau_p$ , where  $\alpha_{EY}(\text{th})=8 \times 10^{-10} \text{ K}^{-2}$ . For the experimental parameters discussed below  $(n_c/n_{\text{imp}})(1/\tau_{EY})$  is three orders of magnitude smaller than the leading contributions to  $1/T_2^*$ . Thus, to simplify our analysis of experiments on  $n$ -GaAs, we set  $\alpha_{EY}=0$  and  $1/\tau_c=1/\tau_{DP}$ .

Spins localized on donor sites cannot relax by the same scattering-dependent processes that relax conduction-band spins. As a rule, relaxation times for localized states are longer than for itinerant states, due to phase-space effects. Lifetimes for localized states can be very long: times in excess of  $10^3$  s have been measured for donor bound states of phosphorus-doped silicon.<sup>12</sup>

**Spin-phonon Mechanism.** Acoustic phonons can relax localized spins by dephasing, due to spin-orbit coupling mixing spin states, if an external field  $B$  breaks the time-reversal symmetry present at zero field (Van Vleck cancellation). With  $B$  in the  $z$  direction, the relevant term in the Hamiltonian for a single spin takes the form,

$$H_{s-ph}=C\mu_B B\sigma_{zi}\Delta_i, \quad (10)$$

where  $\Delta_i$  is the dilatation at a donor site  $i$  and  $C$  is a constant. The dilatation modulates  $g^*$ , the effective  $g$  factor, which is given by  $g^*=2[1-(m^*/m-1)\Delta_{so}/(3E_g+2\Delta_{so})]$ , where  $E_g=1.4$  eV is the energy gap,  $m^*/m=0.067$  is the ratio of the effective mass to the bare mass, and  $\Delta_{so}=0.344$  eV is the spin-orbit splitting of the valence bands.<sup>13</sup> We also have that  $E_g(\Delta_i)=E_g(0)-(9 \text{ eV})\Delta_i$  and the effective mass depends on the gap as  $m^* \sim 1/E_g$ . These facts allow us to estimate  $C^2 \approx 550$ . There is no generally accepted theory of the multispin relaxation rate  $1/T_2^*$  that results from such a Hamiltonian (the single-spin rate  $1/T_2$  has recently been calculated in silicon<sup>14</sup>). We can obtain a simple estimate using Redfield theory, which yields  $1/\tau_{s-ph}=C^2B^2\langle\Delta^2\rangle\tau_{ph}$ , where  $\tau_{ph}$  is the phonon correlation time<sup>15</sup> and  $\Delta$  is the average dilatation for the occupied donors. This leads finally to  $1/\tau_{s-ph}=\alpha_{s-ph}B^2T^4f(T)$  where  $f(T)=\int_0^{\theta_D/T} x^3[(1/2)+(e^x-1)^{-1}]dx$  in a Debye model for the phonons and  $\theta_D=343$  K for GaAs. We note that this theory is not likely to be valid at higher temperatures, where multiphonon and Orbach processes become important. This issue is not settled even in insulators, and we defer full consideration of it to a later publication.

**Nuclear Hyperfine Mechanism.** A localized electron

spin can relax through the hyperfine interaction with the  $N \approx 10^5$  nuclei with which it is in contact. The nuclei are randomly oriented under most conditions, and the associated field felt by the electron is  $A/\sqrt{Ng}\mu_B \approx 10^{-2}$  T, where  $A$  is the hyperfine constant. The corresponding precession frequency is  $\omega_N \approx 10^8 \text{ s}^{-1}$ . There is also a characteristic time for the nuclei  $T_{n2} \approx 10^{-4}$  s, which comes from the nuclear dipole-dipole interaction. Hence,  $\omega_N T_{n2} \approx 10^4$ , and we are in the regime where the effective random field fluctuates slowly compared to the precession of the spin. The relaxation time from coupling to the nuclei,  $1/\tau_{nuc}$ , is not expected to have strong temperature dependence in the range  $T > 1$  K considered here, and there should be no field dependence as long as  $B > 10^{-2}$  T. We treat  $1/\tau_{nuc}$  as a constant.

**Dzyaloshinskii-Moriya Mechanism.** Localized electron spins can relax by the Dzyaloshinskii-Moriya (DM) interaction.<sup>16</sup> This interaction, arising from spin-orbit coupling, produces a term proportional to  $\vec{b} \cdot \vec{s}_1 \times \vec{s}_2$ , where  $\vec{b}$  is related to the interspin separation and to the exchange integral between the wave function on sites 1 and 2. This interaction is not isotropic in spin space and can therefore relax the spins. The calculation of the effect of this term on  $1/T_2^*$  is not straightforward, since it involves aspects of the spin-glass problem that are not entirely solved. Gor'kov and Krotkov<sup>17</sup> have given the first term in a density expansion. We use their expression, though with a more general distance dependence for the exchange interaction,<sup>18</sup> as a first step toward a theory valid at higher impurity densities. We find  $1/\tau_{DM}=\alpha_{DM}n_{\text{imp}}\alpha_B^3f_{DM}(T)$ . Here  $\alpha_{DM}(\text{th})=0.01 \text{ ns}^{-1}$  and the weakly temperature-dependent, dimensionless function  $f_{DM}(T) \approx 32$  at  $T=5$  K.

The total relaxation rate for the localized spins in our theory is given by  $1/\tau_l=1/\tau_{s-ph}+1/\tau_{nuc}+1/\tau_{DM}$ .

In Figs. 1 and 2 we compare the results of our theory to experimental data on  $n$ -GaAs at  $n_{\text{imp}}=10^{16} \text{ cm}^{-3}$  and  $n_{\text{imp}}=10^{18} \text{ cm}^{-3}$ . Our procedure is as follows. Each mechanism above has very definite field, temperature, and doping dependence. The overall constant factor for each is less certain. The data are consistent with  $1/\tau_{nuc}=0$ . However, for reasons explained below, the data do not set tight limits on  $1/\tau_{nuc}$ . We do a least-squares fit to the complete data set shown in Figs. 1 and 2, using the three remaining adjustable parameters, with the results  $\alpha_{DP}(\text{exp})=8.6 \times 10^{-10} \text{ K}^{-3} \text{ ps}^{-2}$ ,  $\alpha_{s-ph}(\text{exp})=2.2 \times 10^{-11} \text{ T}^{-2} \text{ K}^{-4} \text{ ns}^{-1}$ , and  $\alpha_{DM}(\text{exp})=0.031 \text{ ns}^{-1}$ . These values are in satisfactory agreement with  $\alpha_{DP}(\text{th})=9.0 \times 10^{-10} \text{ K}^{-3} \text{ ps}^{-2}$  and  $\alpha_{DM}(\text{th})=0.01 \text{ ns}^{-1}$ . The remaining value  $\alpha_{s-ph}(\text{exp})$  is best viewed as an estimate of the phonon correlation time,  $\tau_{ph}=1.2 \times 10^{-5} \text{ ns}$ . This seems reasonable, given that the inverse Debye frequency  $\hbar/k_B\theta_D=2.2 \times 10^{-5} \text{ ns}$ . But true comparison of theory and experiment for this prefactor awaits a more comprehensive theory of the phonon relaxation, as noted above.

The fits against temperature at  $n_{\text{imp}}=10^{16} \text{ cm}^{-3}$  for  $B=0$  T and 4 T are shown in Fig. 1. There are two surprising points about the data: (1)  $1/T_2^*$  is independent of  $B$  at high  $T$ ; (2) the  $T$  dependence is nonmonotonic at higher fields. Point (1) is explained by noting that in our theory all  $B$  dependence comes from  $1/\tau_{s-ph}$ . For this doping, the localized states are completely depopulated at high  $T$ . Point (2) is more subtle.

In our theory, though both  $1/\tau_c$  and  $1/\tau_l$  increase with increasing  $T$ , their contributions to  $1/T_2^*$  are weighted by  $n_c/n_{imp}$  and  $n_l/n_{imp}$ , respectively. Starting at low  $T$ , where localized states dominate, we have  $1/T_2^*$  decreasing with increasing  $T$ , due to the decrease of  $n_l/n_{imp}$  as  $T$  increases. After the localized states are depopulated  $1/\tau_c$  dominates and  $1/T_2^*$  is an increasing function of  $T$ .

The fits against field for  $n_{imp}=10^{16}$  cm $^{-3}$ , and  $n_{imp}=10^{18}$  cm $^{-3}$  at  $T=5$  K are shown in Fig. 2. These results are also surprising: there is a strong enhancement of  $1/T_2^*$  by  $B$  at low doping, while at high doping, the dependence is quite weak. In this case, the explanation relies on the DM contribution. At high doping  $1/\tau_{DM}$  dominates, because of the short impurity-impurity spacing and consequent fast relaxation. This rate is field independent. At low doping  $1/\tau_{s-ph}$ , with its strong  $B$  dependence, is more important.

Experiments<sup>3,4</sup> show  $n_{imp}=10^{16}$  cm $^{-3}$  is the “optimal” (smallest  $1/T_2^*$ ) doping value at low  $T$ . Going to lower dopings (data not shown) increases  $1/T_2^*$ . This is due to a combination of effects. Increasing the doping at first raises the number of localized states, which have intrinsically much smaller decay rates. However, this process stops when the impurity states begin to overlap, and  $1/\tau_{DM}$  dominates. This is why the minimum  $1/T_2^*$  is near the metal-insulator transition. These conclusions are consistent with those of Ref. 4 at small  $B$  and  $T$ .

From an examination of Fig. 2, we can see why adding  $1/\tau_{nuc}$  does not significantly improve or worsen the fit. Doing so adds a constant to  $1/T_2^*$ , which would move all theory curves rigidly upward, not much affecting the overall good-

ness of fit. Thus, we cannot get a meaningful limit on  $1/\tau_{nuc}$  with this data set. The question of how to average over nuclear degrees of freedom to find  $1/\tau_{nuc}$  is not completely clear, and there are different results in the recent literature.<sup>19,20</sup>

The main shortcoming in our theory lies in the high-doping regime, where spin-glass effects become important. This is shown by the  $B=0$  points in Fig. 2, where  $1/T_2^*$  is dominated by  $1/\tau_{DM}$ . Since we use a low-density (pairwise correlations only) expansion for  $1/\tau_{DM}$ , the ratio of the theory points is exactly the ratio between the densities, which is too small compared with experiments. A better theory would approach freezing as a collective, not pairwise, effect. Apart from this, all the qualitative features of the data are explained in our picture.

Our analysis shows that the complicated  $B$  and  $T$  dependencies for  $1/T_2^*$  observed in experiments are due to having two strongly interacting subsystems of spins: one localized and the other itinerant. The coupling of localized spins to phonons gives rise to the unusual magnetic-field dependencies of the relaxation rates. Overall, the very different  $T$  and  $B$  dependencies for  $1/\tau_c$  and  $1/\tau_l$  coupled with population effects give the wide range of experimental phenomena observed.

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