

Phonon-Induced Spin Relaxation of Conduction Electrons in Aluminum

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(Received 12 April 1999)

Spin-flip Eliashberg function $\alpha_S^2 F$ and temperature-dependent spin relaxation time $T_1(T)$ are calculated for aluminum using realistic pseudopotentials. The spin-flip electron-phonon coupling constant λ_S is found to be 2.5×10^{-5} . The calculations agree with experiments validating the Elliott-Yafet theory and the spin-hot-spot picture of spin relaxation for polyvalent metals.

PACS numbers: 71.70.Ej, 75.40.Gb, 76.30.Pk

Spin dynamics of itinerant electrons in metals and semiconductors is attracting increasing attention. Part of the reason for this interest is fundamental, arising from improved spin injection and detection techniques [1] which now allow precise measurements of spin transport, relaxation, and coherence properties. But much of the recent interest is also motivated by the exciting potential of using electron spin as a building block in nanoelectronics (dubbed “spintronics”) where spin dynamics and transport is projected to be utilized in proposed novel device applications. The most ambitious such possibility is using electron spin as a qubit in a quantum computer architecture, but more modest proposals involving the use of spin injection and transport in new quantum transistor devices (“spin transistors”) have also been made [1].

Electron spin already plays a fundamental, albeit passive, role in giant magnetoresistance-based memory devices. The current push for a better understanding of spin dynamics in electronic materials is, however, based on the hope that the electron spin could be used as an *active* element, where manipulation of spin in a controlled manner will lead to novel device applications which are not feasible in conventional microelectronics. This hope arises from two underlying concepts: the inherently quantum mechanical nature of spin (enabling the possibility of truly quantum devices which could not be envisioned within standard micro- or nanoelectronics), and, even more importantly, the inherently long relaxation or coherence time of spin eigenstates in metals and semiconductors (indeed, in a typical nonmagnetic metal at room temperature electron spins survive for hundreds of picoseconds; by comparison, momentum states live no more than femtoseconds). This Letter provides the first realistic quantitative calculation of the temperature dependent spin relaxation time (the so called T_1 relaxation time) in an electronic material, namely, metallic aluminum. The calculation, for reasons to be explained below, is surprisingly subtle and extremely computationally demanding; it has therefore never been attempted before, although the basic theory for the phenomenon goes back more than thirty-five years [2,3].

The mechanism behind spin relaxation in metals is believed to be the spin-flip scattering of electrons off

phonons and impurities, as suggested by Elliott [2] and Yafet [3]. There are two physical processes to be considered. (A) The periodic, ion-induced spin-orbit interaction is modified by phonons [4]. Electrons scattering by the modified interaction can directly change their spin states. (B) Because of the spin-orbit interaction electronic Bloch states have both spin up and spin down amplitudes. The states can still be polarized by a magnetic field (so we can call them up and down) but because of the spin mixing, even a spin-independent interaction with phonons or impurities (which are assumed to be nonmagnetic) leads to a transition from, say, up to down, degrading any unbalanced spin population. (Note that the spin-orbit interaction by itself does not produce spin relaxation—what is needed is spin-orbit coupling to mix the up and down spins, and a momentum conservation-breaking mechanism such as impurities or phonons.) Although the above processes seem to provide a consistent picture of experimental findings, there has been to date no calculation of T_1 for a metal based on the Elliott-Yafet theory.

In this Letter, we calculate the phonon contribution to T_1 for aluminum providing the first quantitative justification of the theory. (Impurities in real samples contribute only a temperature independent background which can be subtracted from the measurement.) At temperatures T above 100 K, where experimental data are not available, our calculation is a prediction which should be useful for designing room-temperature spintronic devices that use aluminum. We also calculate the spin-flip Eliashberg function $\alpha_S^2 F(\Omega)$ which measures the ability of phonons with frequency Ω to change electron momenta *and* spins. This function, which is an analog of the ordinary (spin-conserving) Eliashberg function $\alpha^2 F(\Omega)$ [5], is important in spin-resolved point-contact spectroscopy where phonon-induced spin flips could be directly observed. (A recent effort [6] to detect phonon-induced spin flips in aluminum failed because of the overwhelming spin-flip boundary scattering in the sample.)

Aluminum belongs to the group of metals whose spin relaxation is strongly influenced by band-structure anomalies [7]. Monod and Beuneu [8] observed that while simple estimates based on the Elliott-Yafet theory work well for monovalent alkali and noble metals, they severely

underestimate $1/T_1$ for polyvalent Al, Mg, Be, and Pd (the only polyvalent metals measured so far). Silsbee and Beuneu [9] pointed out that in aluminum accidental degeneracies can significantly enhance $1/T_1$. We recently [7] developed a general theory including band structure anomalies like accidental degeneracy, crossing Brillouin zone boundaries or special symmetry points, and rigorously showed that they all enhance $1/T_1$. This explains the Monod-Beuneu finding because the anomalies (which we named “spin hot spots” [7]) are ubiquitous in polyvalent metals. The present calculation is consistent with the spin-hot-spot picture. We note that spin hot spots affect only processes (B) of the Elliott-Yafet mechanism so that (A) are negligible for polyvalent metals except at very low temperatures. Processes (A), however, should be more relevant for monovalent alkali and noble metals. Our calculation includes both (A) and (B).

The formula for the spin relaxation rate, first derived by Yafet [3], can be written in the more conventional electron-phonon terminology [5,10] as

$$1/T_1(T) = 8\pi T \int_0^\infty d\Omega \alpha_S^2 F(\Omega) \frac{\partial N(\Omega)}{\partial T}, \quad (1)$$

where $N(\Omega) = [\exp(\hbar\Omega/k_B T) - 1]^{-1}$ and $\alpha_S^2 F(\Omega)$ is the spin-flip Eliashberg function. Before writing the expression for $\alpha_S^2 F$ we introduce the following notation. Electron states Ψ (normalized to a primitive cell) in the periodic potential V containing the spin-orbit interaction are labeled by lattice momentum \mathbf{k} , band index n , and spin polarization \uparrow or \downarrow . If V has inversion symmetry (as in aluminum), states $\Psi_{\mathbf{k}n\uparrow}$ and $\Psi_{\mathbf{k}n\downarrow}$ are degenerate [2]. The spin polarization then means that these two states are chosen to satisfy $(\Psi_{\mathbf{k}n\uparrow}, \hat{\sigma}_z \Psi_{\mathbf{k}n\uparrow}) = -(\Psi_{\mathbf{k}n\downarrow}, \hat{\sigma}_z \Psi_{\mathbf{k}n\downarrow}) > 0$ with the off-diagonal terms vanishing [3]. Lattice vibrations are represented by phonons with momentum \mathbf{q} and polarization index ν . Phonon frequency is $\omega_{\mathbf{q}\nu}$ and polarization vector $\mathbf{u}_{\mathbf{q}\nu}$ (we consider a Bravais lattice). If $\mathbf{q} = \mathbf{k} - \mathbf{k}'$ and

$$g_{\mathbf{k}n\uparrow, \mathbf{k}'n'\downarrow}^\nu \equiv |\mathbf{u}_{\mathbf{q}\nu} \cdot (\Psi_{\mathbf{k}n\uparrow}, \nabla V \Psi_{\mathbf{k}'n'\downarrow})|^2, \quad (2)$$

the spin-flip Eliashberg function is

$$\alpha_S^2 F(\Omega) = \frac{g_S}{2M\Omega} \sum_\nu \langle\langle g_{\mathbf{k}n\uparrow, \mathbf{k}'n'\downarrow}^\nu \delta(\omega_{\mathbf{q},\nu} - \Omega) \rangle\rangle_{\mathbf{k}n} \rangle_{\mathbf{k}'n'}. \quad (3)$$

Here g_S is the number of states per spin and atom at the Fermi level, M is the ion mass, and $\langle\langle \dots \rangle\rangle_{\mathbf{k}n}$ denotes the Fermi surface averaging [11].

We calculate $\alpha_S^2 F$ and T_1 for aluminum by the pseudopotential method [5]. The spin-independent part of the electron-ion pseudopotential is represented by the Mašović-Zeković [12] semiempirical form factor which reproduces well the observed band gaps at the symme-

try points of the Brillouin zone. This is a crucial feature because the presence of spin hot spots makes T_1 sensitive to the band structure at the Fermi surface [7]. The spin-orbit part of the pseudopotential comes from a fit of the first-principles Bachelet-Hamann-Schlüter pseudopotential [13] to $\alpha \hat{\mathbf{L}} \cdot \hat{\mathbf{S}} \mathcal{P}_l$, where $\hat{\mathbf{L}}$ ($\hat{\mathbf{S}}$) is the orbital (spin) momentum operator and \mathcal{P}_l is the operator projecting on the orbital momentum state l . The parameter $\alpha = 2.4 \times 10^{-3}$ a.u. (1 a.u. = 2 Ry) inside the ion core of twice the Bohr radius, $r_c = 2r_B$. Outside the core α vanishes. The cutoff for the plane-wave energy is 1 a.u. from the Fermi level [14]. For phonons we use the highly successful force-constant model of Cowley [15] which gives an excellent fit to the experimental spectrum. Finally, the sums over the Brillouin zone are done by the tetrahedron method [16] with a specially designed grid of more than 4000 points around the Fermi surface in an irreducible wedge of the Brillouin zone to accurately obtain contributions from the spin hot spots.

Figure 1 shows the calculated spin relaxation time T_1 as a function of temperature. The agreement with experiment is evident. At high temperatures where there are no experimental data, our calculation predicts $T_1[\text{ns}] \approx 24 T^{-1}[\text{K}^{-1}]$. This behavior is expected for a phonon-induced relaxation above the Debye temperature which for aluminum is about 400 K. As Fig. 1 shows, the $T_1 \sim T^{-1}$ behavior starts already at 200 K. At very low temperatures the theory predicts the asymptotic temperature dependence $T_1 \sim T^{-5}$ (the Yafet law [3]) purely on dimensional grounds. Our calculation gives rather a good fit to $T_1 \sim T^{-4.35}$ between 2 and 10 K. At lower temperatures our results cease to be reliable because of the finite size (limited by the computing resources) of the tetrahedron blocks in the summations over the Brillouin

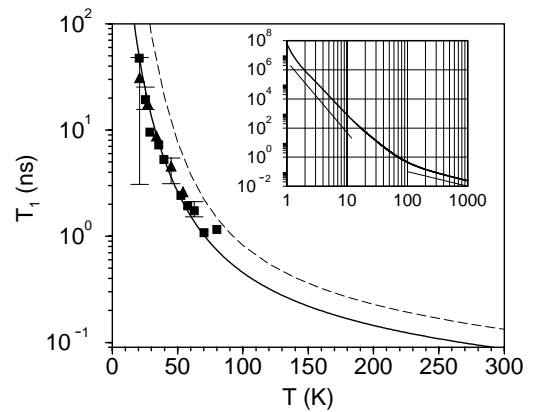


FIG. 1. Calculated spin relaxation time T_1 of conduction electrons in aluminum as a function of temperature T (solid line). Symbols are experimentally determined [17] phonon contribution to T_1 from measurements by Johnson and Silsbee [17] (triangles) and Lubzens and Schultz [18] (squares). The dashed line is an estimate of T_1 from Eq. (4). The inset shows T_1 over a wider temperature range with thin lines indicating the predicted low- T ($T_1 \sim T^{-5}$) and high- T ($T_1 \sim T^{-1}$) behavior.

zone. We anticipate that the asymptotic Yafet law would be reached at lower temperatures (much lower than 2 K) since we have verified numerically its origin, namely that $g_{\mathbf{k}n\mathbf{l},\mathbf{k}'n\mathbf{l}}^v \sim (\mathbf{k} - \mathbf{k}')^4$ at $\mathbf{k} \rightarrow \mathbf{k}'$ [3] (a quadratic dependence would be expected for spin-conserving matrix elements). In Fig. 1 we also plot an estimate of T_1 based on the simple formula [7]

$$T_1 \approx \tau/4\langle b^2 \rangle, \quad (4)$$

where $\langle b^2 \rangle$ is the Fermi surface average of the spin-mixing parameter, calculated in [7] to be 2.5×10^{-5} , and τ is the momentum relaxation time obtained from the Drude formula for the resistivity (resistivity data taken from Ref. [19]) with an electron thermal mass of 1.5 [20] of the free electron mass. This estimate of T_1 reproduces well the calculated functional temperature dependence, making Eq. (4) useful as a starting point for order-of-magnitude estimates.

The calculated spin-flip Eliashberg function $\alpha_S^2 F$ for aluminum is shown in Fig. 2 along with the phonon density of states F and the spin-conserving Eliashberg function $\alpha^2 F$. The last agrees very well with previous calculations [21,22]. Transverse phonon modes which dominate the low-frequency spectrum are less effective in scattering electrons, with or without spin flip, than high-frequency longitudinal phonon modes. The behavior of $\alpha_S^2 F$ at small Ω that gives the Yafet law is predicted to be $\alpha_S^2 F \sim \Omega^4$. We are not able to reproduce this result, again because of the finite size of the tetrahedron blocks. This is a well-known problem that the asymptotic low-frequency behavior is hard to reproduce [21,22].

From the Eliashberg function we can calculate the effective electron-phonon coupling constant

$$\lambda_{(S)} = 2 \int_0^\infty \frac{d\Omega}{\Omega} \alpha_{(S)}^2 F(\Omega). \quad (5)$$

We obtain $\lambda \approx 0.4$ and $\lambda_S \approx 2.5 \times 10^{-5}$. The spin-conserving λ falls well into the interval of the

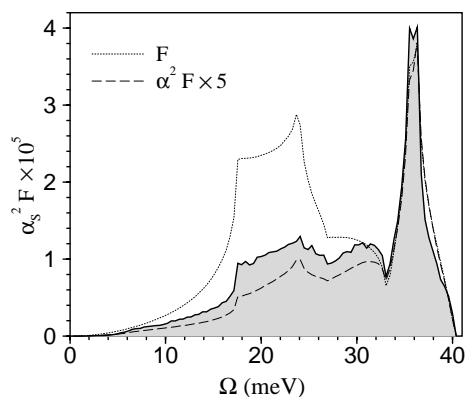


FIG. 2. Spin-flip Eliashberg function $\alpha_S^2 F$ for aluminum. The dotted line shows the phonon density of states F and the dashed line is the ordinary (spin-conserving) Eliashberg function $\alpha^2 F$. The curves are calculated for the model described in the text.

“recommended” values 0.38–0.48 [5] obtained by different methods [21–24]. At high temperatures the phonon-induced relaxation is determined by $\lambda_{(S)}$, since in this regime $\hbar/\tau \approx 2\pi\lambda k_B T$ and $\hbar/T_1 \approx 4\pi\lambda_S k_B T$. The momentum to spin relaxation time ratio τ/T_1 is $2\lambda_S/\lambda \approx 1.24 \times 10^{-4}$. From the above ratio of τ/T_1 we obtain the “effective” $\langle b^2 \rangle \approx 3.1 \times 10^{-5}$ in Eq. (4), not that different from its calculated value of 2.0×10^{-5} [7]. Thus, our theory is internally consistent.

We conclude with a remark on the accuracy of our calculation of λ_S . The numerical error is accumulated mostly during the summations over the Brillouin zone. This error was previously estimated [22] to be about 10%. Another source of uncertainty, which is much more important here than in the spin-conserving calculations, comes from the choice of the pseudopotentials. While the spin-orbit pseudopotential sets the overall scale ($1/T_1 \sim \alpha^2$), the scalar part of the pseudopotential determines the “band renormalization” of $1/T_1$, that is, the enhancement due to spin hot spots [7]. Here we can only offer a guess. Considering the spin-orbit part “fixed,” our semiempirical scalar pseudopotential, which reproduces the experimental band gaps at symmetry points within 5%, does not introduce more than another 10% error [25], making λ_S determined with 20% accuracy. As for the spin-orbit interaction, future experiments done in the regime where $T_1 \sim 1/T$ (that is, above 200 K), will have the opportunity to set definite constraints on α through a direct comparison with our theory.

In summary, we have provided the first fully quantitative theory for the temperature-dependent spin relaxation rate in aluminum taking into account spin-orbit coupling and electron-phonon interaction within the Elliott-Yafet formalism using realistic pseudopotentials. Our theoretical results are in excellent agreement with the measured $T_1(T)$ in aluminum and for $T > 100$ K, where experimental results are currently nonexistent, our theory provides specific predictions for comparison with future experiments.

We thank P.B. Allen for helpful discussions. This work was supported by the U.S. ONR and the U.S. ARO.

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