PHYSICAL REVIEW B VOLUME 35, NUMBER 13 1 MAY 1987

# Relaxation of nuclear and electronic magnetic moments in heavy-electron compounds

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(Received 20 June 1986)

Estimates are presented of nuclear-spin-lattice relaxation rates in the normal state of heavy-electron metals due to dipolar coupling of nuclear moments and $4f$ or $5f$ spin fluctuations. The $4f$ or $5f$ sites are modeled as point ions in the $LS$ limit with dynamics of the $N_{\mathrm{grd}}$-fold degenerate Anderson model. Dipolar coupling alone is sufficiently large to account for the relaxation of $^9$Be in CeBe$_{13}$ and UBe$_{13}$, as has been found for other light nuclei in heavy-electron and local moment systems. However, for heavy nuclei with large intrinsic relaxation rates due to stronger on-site coupling to conduction electrons, the dipolar contribution underestimates the observed relaxation by orders of magnitude. I suggest that nuclear resonance is a more effective probe than electron-spin resonance of the heavy-fermion state primarily because the probe nuclei sit closer to the heavy-electron sites so that the strong range dependence of both dipolar and transferred exchange (Ruderman-Kittel-Kasuya-Yosida) interactions favors nuclear relaxation. I show for the Anderson lattice, using both mean field formalism and Green's function formalism (the latter explicitly including dynamic many-body effects and assuming a dispersionless interaction contribution to the $f$-electron self-energy) that many-body cancellation effects are relevant in magnetic relaxation in heavy-electron compounds only when coupling of probe moments to distant heavy-electron sites is vanishingly small. I show that since the maximal $f$-electron enhancement of impurity electronic moment relaxation follows the square of the ratio of the paramagnetic transition temperature ($T_P$) to the effective degeneracy temperature or "Kondo" energy scale ($T_0$) that those heavy-fermion systems close to magnetic instabilities are more suitable for study with electron spin resonance. Detailed studies of the relaxation in that instance are beyond the scope of this paper.

## INTRODUCTION

Heavy-electron systems have attracted much attention recently, both for the onset of apparently exotic superconductivity in CeCu$_2$Si$_2$, UPt$_3$, and UBe$_{13}$ as well as the spectacularly enhanced normal state specific heat and magnetic susceptibility.[1,2] The understanding of these materials is very difficult as is evidenced by the experimental situation in magnetic resonance.[3] For example, in UBe$_{13}$, nuclear magnetic resonance (NMR) on the $^9$Be nuclei shows a low-temperature spin-lattice relaxation rate ($1/T_1$) enhanced by a factor of 1000 over the rate in LaB$_{13}$ or Be metal.[4] The temperature dependence of the relaxation follows that expected from a narrow resonance of width $\sim 10$ K pinned to the Fermi level as is also suggested by the electronic specific heat.[5]

While this rate enhancement is indeed enormous, it falls far short of the $10^6$-fold enhancement one would expect by comparing the low-temperature electronic specific heat of UBe$_{13}$ to LaBe$_{13}$, and assuming as per the standard Korringa law for magnetic relaxation in metals that $(T_1T)^{-1} \sim N^2(0)$, $N(0)$ being the density of states at the Fermi level, proportional to the specific-heat coefficient. Moreover, while NMR on the $^9$Be nuclei clearly shows enhanced relaxation, electron-spin resonance (ESR) on Er and Dy substituted for U shows only a moderate temperature enhancement of spin-lattice relaxation rates by a factor of 2–3.[6] Conceptually, these experiments do not differ in any apparent gross features. Despite the intriguing suggestion for ESR that many-body mass enhancements cancel from $1/T_1$,[7] the NMR and ESR results appear to be at odds. It should be noted that in CePd$_3$ and CeOs$_2$ which are generally classified as valence fluctuation materials (corresponding to the interpretation in this paper of having large characteristic or Kondo temperatures), the impurity ESR relaxation appears to be suppressed relative to isostructural La-based analogues.[8–10] However, it is generally the case that the impurity Korringa rates are unenhanced or mildly enhanced over those in the noninteracting hosts.[11,12] A possible exception to this is Gd in UAl$_2$ for which no ESR signal has been observed, due either to an anomalously large $g$ shift or highly enhanced relaxation.[12]

In this paper I point out a possible explanation for the observed nuclear relaxation rates in UBe$_{13}$ and CeBe$_{13}$, some of the reasons why ESR is generally a less effective probe of the heavy-electron state than NMR, and how the simplest inclusion of lattice coherence effects due to the *banding* of the heavy electrons do not alter this picture substantially. The basic physical idea is that the heavy-electron magnetic moments reside at the $4f$ or $5f$ sites rather distant from the probe NMR or ESR moments. For light nuclei, direct coupling via the dipolar interaction is sufficient to provide strong relaxation due to the fluctuating $4f$ or $5f$ moments. For heavy nuclei and impurity electronic moments the dominant coupling should be via conduction-electron-mediated [Ruderman-Kittel-Kasuya-Yosida (RKKY)] interactions which will likely be weak compared to on-site coupling of the conduction-electron spins. Indeed, due to the physical reality that *f*-*f* separations in these intermetallic compounds are larger than that between the *f* and nuclear resonance probe sites, the chances of any ESR enhancement via coupling to the *f* electrons are small.

The paper is divided into two parts. The first discusses the dipolar mechanism in detail, and the second discusses the cancellation effects and the differences between ESR and NMR. The second section relies upon Anderson lattice formalism the structure of which is readily apparent within recently developed mean field theories. These results should be reasonable so long as intersite spin correlation effects are small and in that limit lead to essentially the same conclusions arrived at in the first section. The central results of this second section with little essential modification also derive from a Green's function treatment (which explicitly includes dynamical many-body effects) with the assumption of an *f*-electron self-energy which has dispersion only through one-body hybridization effects. The contribution from electronic interactions is assumed frequency dependent only. Details of this derivation are deferred to an appendix. The paper includes two tables. Table I summarizes the relevant interactions which can lead to relaxation in these materials, and Table II summarizes the results of the dipolar rate estimates. Note that for both NMR and ESR considerations, I suppose that the relaxation rate of the heavy-electron moments is large compared to that of the probe moment.

## DIPOLAR COUPLING TO LIGHT NUCLEI

In an effort to clarify the physics of the problem, I have estimated the nuclear relaxation rates induced by dipolar coupling of 4*f* or 5*f* spin fluctuations to the nuclear moment. This coupling mechanism represents a clear difference between ESR and NMR, since electronic dipole coupling is not expected to have sufficient strength (relative to electronic exchange energies) to be relevant.[13] Note that the various relevant interactions for the problem are summarized in Table I. For the purposes of this paper, it is sufficient to limit considerations to crystals with cubic symmetry at the 4*f* or 5*f* sites, and to compute powder averages of the theoretical rates. It is not difficult to extend the calculations beyond these assumptions as the need arises.

In performing the calculations, I have assumed (i) that the 4*f* or 5*f* ions possess point moments; (ii) that the $N_{\mathrm{grd}}$-fold degenerate lowest-lying multiplets are described by Hund's rules; and (iii) that the $N_{\mathrm{grd}}$-fold degenerate Anderson model in the limit of unit occupancy adequately describes the dynamics.[14]

We expect (i)–(iii) above to be very reasonable for Ce compounds to the extent that intersite correlations are negligible. However, for U compounds with less localized 5*f* electrons, (i) will surely not hold, though one would not expect corrections to create an order of magnitude difference. Point (ii) could be objected to more seriously, since the strong spin-orbit coupling in the actinides is known to require an intermediate-coupling description. However, some calculations appear to yield results closer to *LS* coupling than the extreme *jj* coupling.[15] Finally, point (iii) could be objected to since a more complicated Anderson model with $5f^1$, $5f^2$, and $5f^3$ configurations might be envisaged to describe U in a metal host.[16] While this may complicate the actual form of the ground state relative to the simpler case presented by Ce, the basic conclusion of a lowest-lying singlet appears to still come out of the calculations. Moreover, the spectrum of spin fluctuations which depends primarily upon the details of the ground configuration should not be significantly different from the Ce case. In regard to all the above points, since I am interested primarily in order of magnitude estimates of the relaxation rates, I shall press ahead with these admittedly heuristic assumptions.

It should be noted that $1/T_1T$ probes a sum over all sites or equivalently all wave vectors of the dynamic susceptibility.[17] In this sense, it is expected to be less sensitive than neutron scattering to intersite effects which could produce coherent effects in the long-wavelength limit.

While the notion of dipolar coupling to *f*-electron spin fluctuations as a source of strong nuclear relaxation is not new, the present application (arrived at independently of previous work) is new. One of the clearest examples of the dipolar mechanism in the literature is for the local moment system $(\mathrm{Gd},\underline{\mathrm{La}})\mathrm{Al}_2$.[18] However, the heavy-electron systems differ from those studied previously in that the *f*-electron magnetic moments remain strongly damped all the way down to zero temperature. In contrast, the Korringa rate for, say, Gd is orders of magnitude smaller than that of heavy-electron systems for all temperatures and possesses usually only a minute remnant linewidth for $T=0$ K.[19] We note that the high-temperature relaxation of H in $\mathrm{CeH}_2$ (Ref. 20) and of $^{11}\mathrm{B}$ in $\mathrm{CeRh}_3\mathrm{B}_2$ (Ref. 21) are apparently accounted for by dipolar coupling to the *high-temperature* Ce local moments in those systems.

We now proceed to compute the relaxation rate. Following Slichter,[22] the dipolar induced rate for a particular field orientation is written as

$$\left[\frac{1}{T_1}\right]_{\mathrm{dip}}=\frac{3}{2I(I+1)}\frac{2\pi}{\hbar}\times\sum_{i,f}\frac{e^{-\beta E_i}}{Z_nZ_{\mathrm{el}}}\,|\langle i\,|H_{\mathrm{dip}}|\,f\rangle|^2\delta(E_i-E_f)\ . \quad (1)$$

In the above expression, $\beta=1/k_BT$, $I$ is the nuclear spin, $Z_n$ the nuclear partition function, and $Z_{\mathrm{el}}$ is the electronic partition function. $H_{\mathrm{dip}}$ is factored into three terms involving opposing nuclear and electronic spin flips, parallel nuclear and electronic spin flips, and nuclear spin flips with no accompanying electronic spin flips. These are proportional respectively to the spherical harmonics $Y_{21}(\hat{\mathbf{h}})$, $Y_{22}(\hat{\mathbf{h}})$, and $Y_{20}(\hat{\mathbf{h}})$, where $\hat{\mathbf{h}}$ is the direction of the applied dc field. Thus in a powder sample, the dipolar rate can contribute only to the line broadening and not Knight shift of the probe nuclei, since the average over orientations in a powder sample will kill all three of these terms.

The calculation proceeds with substitution of the separate terms of the dipolar interaction into (1) with subsequent factorization of the nuclear and electronic portions of the matrix elements. Use is made of the fact that the nuclear resonance frequencies are negligibly small compared to any typical electronic energies, and averaging is performed over the field direction to provide rate estimates appropriate to a powder sample. I then obtain, for a cubic system in the low-field limit

$$\left\langle \left[ \frac{1}{T_1 T} \right]_{\text{dip}} \right\rangle_{\hat{\text{h}}} = \frac{4k_B}{\hbar} \gamma_N^2 \mu_N^2 \left[ \sum_R \frac{1}{R^6} \right] \lim_{\omega \to 0} \left[ \frac{\chi''(\omega,T)}{\hbar\omega} \right] . \tag{2}$$

Here, $\chi''(\omega,T)$ is the absorptive part of the $f$ electron spin susceptibility, $\gamma_N$ is the nuclear gyromagnetic ratio given by ($h$ is the applied field strength)

TABLE I. Interactions leading to magnetic relaxation of NMR or ESR probe moments in heavy-electron compounds. The table contains nothing new and is included solely for pedagogical purposes. The notation is as follows: $\mathbf{S}_n$ is the probe spin, $\mathbf{S}_c$ is the spin of a conduction electron Wannier orbital about the site, $J_{nc}$ is the probe–conduction-electron exchange, $\mathbf{r}$ is the Wannier orbital coordinate about the probe site ($\hat{\mathbf{r}}=\mathbf{r}/r$), $\psi_c(0)$ is the actual Wannier orbital wave function at the probe nucleus, $l$ is the orbital angular momentum of the Wannier orbital if it is of non-$s$-wave character, $J_{cs}$ is the electrostatic exchange integral between a given conduction electron and an $s$ orbital on-site, $E_s$ is the energy of that $s$ orbital relative to the Fermi level, $\mathbf{J}$ is the total angular momentum of a distant $f$-electron moment at position $\mathbf{R}$ relative to the probe site, $D$ is the conduction bandwidth, $k_F$ is the Fermi momentum, and $J_{cf}$ is the conduction-electron–to–$f$-electron exchange coupling. The formula for the core polarization coupling is rather schematic and ignores mixed exchange between two different core $s$ levels (Ref. 37). The range function $F(k_F R)$ in the RKKY interaction has a nominal asymptotic envelope of $\cos(2k_F R)/R^3$ for a spherical Fermi surface, but of course will depend in detail upon the actual band structure and Fermi surface of the material in question. The bounding of the on-site couplings by a $(Z/n)^3$ envelope ($Z$ the atomic number and $n$ the principle quantum number of the valence electrons) follows by assuming the short-distance behavior to be atomiclike and using hydrogenic orbitals as a crude approximation to this behavior (Ref. 66). In reality, of course, screening of outer orbitals will tend to reduce $Z$ to some effective value depending upon the orbital, and thus the full atomic number must represent some kind of upper bound. The point is that one expects greater overlap at the nucleus for greater atomic number (Ref. 38). Hence, the on-site effects will eventually dominate NMR for large $Z$ with all other electronic and structural properties held constant.

| | NMR | ESR |
|---|---|---|
| | On-site ($J_{nc}$) | |
| Contact exchange<br>$-J_{nc}\mathbf{S}_n\cdot\mathbf{S}_c(0)$ | $s$ wave only<br>$J_{nc}=\frac{8\pi}{3}\gamma_N\mu_N g\mu_B \lvert\psi_c(0)\rvert^2$ | dominant |
| Orbital coupling<br>$\gamma_N\mu_N g\mu_B \frac{\mathbf{S}_n\cdot \boldsymbol{l}_c}{r_{el}^3}$ | non $s$ wave<br>$\sim Z^3$ | |
| Dipolar<br>$\gamma_N\mu_N g\mu_B \frac{\mathbf{S}_n\cdot\mathbf{S}_c-3(\mathbf{S}_n\cdot\hat{\mathbf{r}})(\mathbf{S}_c\cdot\hat{\mathbf{r}})}{r_{el}^3}$ | non $s$ wave<br>$\sim Z^3$ | weaker than exchange |
| Core polarization<br>$-J_{nc}\mathbf{S}_n\cdot\mathbf{S}_c(0)$ | $\lvert J_{nc}\rvert = \left\lvert \sum_s \frac{J_{ns}J_{cs}}{E_s} \lvert\psi_s(0)\rvert^2 \right\rvert$<br>$\sim Z^3$ | |
| | Intersite ($J_{nf}$) | |
| Dipolar<br>$\gamma_N\mu_N g_j\mu_B \frac{\mathbf{S}_n\cdot\mathbf{J}_f-3(\mathbf{S}_n\cdot\hat{\mathbf{R}})(\mathbf{J}_f\cdot\hat{\mathbf{R}})}{R^3}$ | small $Z$ only | weaker than RKKY |
| RKKY<br>$\alpha\frac{J_{nc}J_{nf}}{D}F(R)\mathbf{S}_n\cdot\mathbf{J}_f$ | $F(R)\sim\frac{\cos 2k_F R}{(k_F R)^3}$<br>$\alpha\sim O(1)$<br>not $\mathbf{S}_n\cdot\mathbf{J}_f$ for $N_{\text{grd}}>2$ | not $\mathbf{S}_n\cdot\mathbf{J}_f$ for $N_{\text{grd}}>2$ |

$$\frac{\hbar\omega_{\mathrm{NMR}}}{\mu_N h}=\gamma_N\ , \tag{3}$$

and $R$ indexes the $4f$ or $5f$ positions relative to the probe nucleus. The rapid decay of the dipole summand makes it sufficient for the purposes of our simple estimates to truncate at the first shell of $4f$ or $5f$ ions. Corrections due to further-neighbor shells will in no case considered here make than a 10% enhancement of the estimated rate. I shall ignore the problem of inequivalent sites; for the interesting case of $R\mathrm{Be}_{13}$, where $R$ represents rare-earth atoms, all but one of the 13 Be atoms has two near-neighbor "heavy electron" sites. Hence, the thirteenth site is expected to have little intensity.

While in principle information about $\chi''$ is obtainable through neutron scattering data, I can use the assumption that the Anderson model applies to relate the low-temperature limit of the spin-lattice relaxation rate directly to the linear specific heat coefficient which characterizes the heavy electron behavior. I proceed in two steps. First, the Shiba relation[23] connects $\chi''(\omega,T=0)/\omega$ to the low temperature static susceptibility. For a lowest-lying $N_{\mathrm{grd}}$-fold degenerate multiplet, the relation is

$$\lim_{\omega\to 0}\frac{\chi''(\omega,0)}{\hbar\omega}=\frac{3\pi}{N_{\mathrm{grd}}\mu_{\mathrm{eff}}^2}[\chi(0)]^2\ , \tag{4}$$

where $\chi(0)$ is the zero-temperature static susceptibility and

$$(\mu_{\mathrm{eff}}/\mu_B)^2=\frac{3}{N_{\mathrm{grd}}}\sum_{\alpha}|\langle\alpha|g_J J_z|\alpha\rangle|^2\ , \tag{5}$$

$\alpha$ indexes the states of the ground multiplet and $J_z$ is the $z$ component of the total angular momentum. (We allow for the possibility of crystal-field splitting within a Hund's rule multiplet.) The next step is to use the Wilson ratio to eliminate $\chi(0)$ in favor of the linear specific-heat coefficient $\gamma(0)$ according to[24,25]

$$\chi(0)=\frac{N_{\mathrm{grd}}}{N_{\mathrm{grd}}-1}\frac{3\gamma(0)}{\pi^2 k_B^2}\ . \tag{6}$$

We emphasize that (5) and (6) apply only to the extent that on-site effects dominate the physics and thus application of this analysis to any materials which magnetically order is problematic. I also emphasize that in this limit where intersite correlations are small, (5) and (6) hold only in the limit of extreme unit occupancy. However, this latter point practically excludes only those heavy electron systems with "degeneracy temperatures" of the order of thousands of kelvin, and thus does not present a problem in this context.[26]

We thus are led to the expression

$$\begin{aligned}\lim_{T\to 0}\left\langle\left[\frac{1}{T_1 T}\right]_{\mathrm{dip}}\right\rangle_{\hat{\mathbf{h}}}&=\frac{4\pi}{3}\frac{\mu_N^2\mu_B^2}{\hbar k_B}\gamma_N^2\left[\sum_R\frac{1}{R^6}\right]\left[\frac{N_{\mathrm{grd}}(\mu_{\mathrm{eff}}/\mu_B)^2}{(N_{\mathrm{grd}}-1)^2}\right]\left[\frac{3\gamma(0)}{\pi^2 k_B}\right]^2\\&=6.33\times 10^{-44}\gamma_N^2\left[\frac{N_{\mathrm{grd}}(\mu_{\mathrm{eff}}/\mu_B)^2}{(N_{\mathrm{grd}}-1)^2}\right]\left[\sum_R\frac{1}{R^6}\right]\left[\frac{3\gamma(0)}{\pi^2 k_B}\right]^2\ ,\end{aligned} \tag{7}$$

the latter holding in cgs units. We have applied this formula to several compounds and tabulated the results (Table II). For $\mathrm{CeBe}_{13}$ and $\mathrm{UBe}_{13}$, all estimates are remarkably close to the observed low-temperature rates, the values being rather insensitive to the choice of the lowest multiplet.

It seems likely that at least part of the relaxation of $^{27}\mathrm{Al}$ in $\mathrm{CeAl}_2$ is due to dipolar coupling to the Ce spin fluctuations. However, it is difficult to estimate the appropriate $\gamma(0)$ values for $\mathrm{CeAl}_2$ and $\mathrm{CeB}_6$ because of the presence of low-temperature phase transitions (antiferromagnetic for $\mathrm{CeAl}_2$ and antiferroquadrupolar for $\mathrm{CeB}_6$). I have set bounds by noting (i) that each compound has a quasielastic linewidth $\Gamma_{\mathrm{neut}}$ ($T=0$ K) of order 1 meV or less.[27,28] This sets a lower bound of $\sim 1000$ mJ/mole K$^2$ on $\gamma(0)$ since both empirical correlation plots and Anderson model theory give $\gamma(0)\sim\Gamma_{\mathrm{neut}}(0)$.[29] It should be noted, however, that some of the line broadening is likely due to antiferromagnetic intersite exchange. (ii) In the dilute Ce limit of (Ce, $\underline{\mathrm{La}}$)$\mathrm{Al}_2$ and (Ce, $\underline{\mathrm{La}}$)$\mathrm{B}_6$, physical properties show characteristic (Kondo) temperatures of order 1 K with correspondingly large linear specific heat coefficients of order $10^4$ mJ/(mole Ce) K.[30-32] The extrapolation from dilute to concentrated limits is justified particularly well for Ce in $\mathrm{LaAl}_2$, where near-perfect scaling with Ce concentration is observed for the susceptibility and resistivity at sufficiently high temperatures.[33] Obviously, the neglect of intersite correlations renders these estimates considerably more uncertain than for $\mathrm{CeBe}_{13}$ and $\mathrm{UBe}_{13}$.

It should also be noted that the presence of a sizeable isotropic component to the Knight shift in powder samples of $\mathrm{CeAl}_2$ proves the presence of other coupling mechanisms (presumably RKKY), since the average over orientations of dipolar shifts is zero.[34] The *anisotropic* component of the Knight shift in $\mathrm{CeB}_6$ has been analyzed in terms of the dipolar coupling which appears to provide a perfectly adequate account.[35] The magnitude of the anisotropic components [proportional to $Y_{20}(\hat{\mathbf{h}})$] is of about the same size as the isotropic piece.

One may well ask why any RKKY coupling between the nuclei and $4f$ or $5f$ moments appears to be so irrelevant in the $R\mathrm{Be}_{13}$ systems. The answer goes roughly as follows. Since both RKKY and dipolar interactions fall off nominally as $1/R^3$, and since both must ultimately contain factors of $8\pi\mu_N\mu_B/3$, the dimensionless ratio of

TABLE II. Estimated dipolar contributions to spin-lattice relaxation in various cubic heavy-electron systems. These estimates are based upon Eq. (2). The degeneracy $N_{\mathrm{grd}}$ of the assumed lowest $f$ multiplet is in parentheses beside the state; the nuclear moment $\gamma_N$ (units of $\mu_N$) appears beside the probe nucleus; the nearest-neighbor $f$-site coordination number ($q_N$) appears beside the nearest neighbor $f$-site distance ($a_N$). [The gyromagnetic ratios and structural information can all be obtained from well-known references (Refs. 67 and 68).] As described in the text, the rate estimates include nearest-neighbor $f$ site contributions within a point ion model. The $f$ dynamics are assumed described by the SU($N_{\mathrm{grd}}$) Anderson model (with one exception; see footnote a below). The estimates are expected to work well only for light nuclei (see the preceding table).

| Material | Ground Mult. ($N_{\mathrm{grd}}$) | $\frac{\mu_{\mathrm{eff}}}{\mu_B}$ | $\gamma(0)$ (mJ/mole K$^2$) | Nucleus ($\gamma_N$) | $a(q)$ | $\left\langle \left[ \frac{1}{T_1 T} \right]_{\mathrm{dip}} \right\rangle_{\hbar}$ (1/sec$-K$) |
|---|---|---|---|---|---|---|
| $\mathrm{CeBe}_{13}$ | $4f^1$, $j=\frac{5}{2}$ (6) | 2.54 | 115 (Ref. 69) | $^9$Be (0.78) | 3.0(2) | 0.0029 |
| | $4f^1$, $j=\frac{5}{2}$, $\Gamma_8$ (4) | 1.99 | | | | 0.0034 |
| | $4f^1$, $j=\frac{5}{2}$, $\Gamma_7$ (2) | 1.24 | | | | 0.0057 |
| | expt. (Ref. 52) | | | | | 0.0030 |
| $\mathrm{UBe}_{13}$ | $5f^2$, $j=4$ (9) | 3.58 | 1000 (Ref. 5) | $^9$Be (0.78) | 3.0(2) | 0.25 |
| | $5f^2$, $j=4$, $\Gamma_3$ (2)[a] | 0.00 | | | | $\leq 0.14$ |
| | $5f^3$, $J=\frac{9}{2}$ (10) | 3.62 | | | | 0.23 |
| | $5f^3$, $j=\frac{9}{2}$, $\Gamma_6$ (2)[b] | 2.31 | | | | 1.50 |
| | expt. (Ref. 4) | | | | | 0.19 |
| $\mathrm{CeSn}_3$ | $4f^1$, $j=\frac{5}{2}$ (6) | 2.54 | 53 (Ref. 69) | $^{119}$Sn (2.07) | 3.33(4) | 0.0047 |
| | expt. (Ref. 41) | | | | | 20.0 |
| $\mathrm{CeAl}_2$ | $4f^1$, $j=\frac{5}{2}$, $\Gamma_7$ (2) | 1.24 | $10^3$–$10^4$ (?)[c] | $^{27}$Al (1.44) | 3.34(2)[d] | 0.78–78.0 |
| | expt. (Ref. 34) | | | | | 25.0 |
| $\mathrm{CeB}_6$ | $4f^1$, $j=\frac{5}{2}$, $\Gamma_8$ (4) | 1.99 | $10^3$–$10^4$ (?)[c] | $^{11}$B (2.56) | 3.05(4) | 4.9–490.0 |

[a] Relaxation due to Van Vleck fluctuations, cf. Eq. (8).

[b] $J=\frac{9}{2}$ quartet states depend in detail upon the level splittings (Ref. 42).

[c] Lower bound from quasielastic linewidth, upper via extrapolation from dilute limit; see text for discussion.

[d] Only the three equivalent sites nearest to Ce atoms are included.

the two interactions between a given nucleus and a $f$-electron moment at a distance $R$ is roughly

$$\frac{I(\text{nuclear-electronic dipole})}{I(\text{nuclear-electronic RKKY})} \approx \frac{k_F^3}{|\psi(0)|_{\mathrm{eff}}^2 |J_{cf}/D|} , \quad (8)$$

where $J_{cf}$ is the conduction-electron–$f$-electron exchange, $D$ is the conduction bandwidth, $k_F$ is the Fermi vector, and $\psi(0)_{\mathrm{eff}}$ is the effective overlap of the conduction electron Wannier function at the nuclear site. This overlap is either via direct contact (for $s$-wave conduction electrons), core polarization,[36,37] or direct dipole or orbital coupling (for non-$s$ states).[38,39] In any case, for small atoms, the effective squared wave function is presumably of order $a_0^{-3}$ (assuming that the short-distance behavior of the Wannier orbitals is nearly atomic) and so the product with $k_F^{-3}$ is no more than say 10.[40] The ratio $J_{cf}/D$ is expected to be, say 0.03 or so, so that one would expect, for small atoms, that dipolar coupling should be reasonably strong relative to RKKY. However, for all sources of $\psi(0)_{\mathrm{eff}}$, the overlap will increase with atomic number, with a naive bound set by the hydrogenic result of $(Z/n)^3$ ($Z$ the atomic number and $n$ the principle quantum number of the conduction electrons). Thus the dipolar mechanism can only be relevant in small nuclei.

Indeed, it is clear from Table II that the relaxation of $^{119}$Sn in $\mathrm{CeSn}_3$ (Ref. 41) cannot be accounted for by the dipolar mechanism. This is very plausible because the intrinsic relaxation of the Sn nucleus (as measured in, say, metallic Sn) is already quite large, of order 30/sec at 1 K. Since this reflects the local character of the Sn-electron Wannier functions which I would not expect to change dramatically in the intermetallic host, the on-site coupling induced rate is likely very nearly comparable to that of $\mathrm{CeSn}_3$.

It is worth noting a final possibility for the $\mathrm{UBe}_{13}$ estimate. If the relevant stable $5f$ configuration has two electrons rather than the usually assumed value of three, then the cubic field at the U site can split the $J=4$ ground multiplet with (possibly) a stable $\Gamma_3$ doublet for the ground state.[42] Such a state has vanishing magnetic dipole moment but can couple magnetically to excited $\Gamma_4$ and $\Gamma_5$ triplets. If one *assumes that interactions with conduction electrons provide a broadening* $\Gamma$ for such a level and the presence of an excited $\Gamma_4$ triplet level at energy $\Delta$ with width $\tilde{\Gamma}$, one obtains a dipolar rate of

$$\lim_{T\to 0} \left\langle \left[ \frac{1}{T_1 T} \right]_{\mathrm{dip}} \right\rangle_{\hat{\mathrm{h}}} = 15.3 \frac{k_B}{\hbar} \gamma_N^2 \mu_N^2 \mu_B^2 \left[ \sum_r \frac{1}{R^6} \right] \frac{\tilde{\Gamma}}{\Gamma(\tilde{\Gamma}^2+\Delta^2)} . \quad (9)$$

Note that the factor $\tilde{\Gamma}/(\tilde{\Gamma}^2+\Delta^2)$ is bounded above by $1/2\Delta$. Assuming $\Gamma = 1$ meV (motivated by the specific-heat data of Ref. 5), $\Delta = 15$ meV (motivated by a comparison to the available neutron scattering and Raman spectra)[43,44] we thus obtain the upper bound of 0.14/sec K quoted in the table. More extensive theoretical considerations are underway, and the relevance of this particular model to other properties will be reviewed in a subsequent work.[45]

## CANCELLATION AND COHERENCE EFFECTS

Finally, it is worth making contact with the cancellation arguments[7] put forward previously to explain, among other things, the ESR results for Er and Dy in $\mathrm{UBe}_{13}$. The arguments put forth were based upon general many-body considerations, specifically, the cancellation of wave-function renormalization factors (mass enhancements due to the frequency dependence of the self-energy) in such quantities as the conductivity, ultrasonic attenuation amplitude, and spin-lattice relaxation. I do not dispute the applicability of these arguments to the first two cases where they may indeed be relevant. Also, as pointed out in the article, the cancellation arguments work only to the extent that "scattering of conduction electrons" provides the dominant coupling to the nuclear or impurity electron moments. For example, in the case of $\mathrm{CeSn}_3$, since the effective on-site coupling is quite large, one can expect the cancellation arguments to apply reasonably well. Independently, a similar viewpoint has been put forth,[46] though the arguments which follow provide a more detailed picture of the physics of the problem.

Such arguments can be put on a particularly transparent basis via the one-body resonant level lattice model which has emerged from various mean-field treatments as a description of the Anderson lattice in the low temperture hydrodynamic regime.[47–49] The effective Hamiltonian is

$$H = \sum_{k,\sigma} [\varepsilon_k c^\dagger_{k\sigma} c_{k\sigma} + \varepsilon_r f^\dagger_{k\sigma} f_{k\sigma} + \tilde{V}(c^\dagger_{k\sigma} c_{k\sigma} + \mathrm{H.c.})] \; . \tag{10}$$

Here, $c^\dagger_{k\sigma}$ creates a normal electron quasiparticle residing in a band of width $D$ of wave vector $k$ and spin $\sigma$, and $f^\dagger_{k\sigma}$ creates a heavy ($f$-electron) quasiparticle. It is assumed that $\varepsilon_r > \tilde{V}^2/D \sim T_0/N_{\mathrm{grd}}$, where $T_0$ is the one-ion Kondo temperature. (Note that the tacit assumption of $N_{\mathrm{grd}}$ fold degenerate *conduction* states is really a theoretical convenience here and consequently I stress only the qualitative aspects of normal conduction electron relaxation of magnetic moments in what follows. Obviously this sidesteps the question of how many bands form, but in the absence of gaps or pseudogaps in the spectra at the Fermi level, I would expect the model to be qualitatively reliable.) Assuming no $k$ dependence to the hybridization $\tilde{V}$, then the mass enhancements are due to wave-function renormalization, as assumed in the paper concerning cancellation arguments. It has been demonstrated previously that the mass factors explicitly cancel in a calculation of the electrical conductivity for this model.[50] Note that while a full calculation should include fluctuation corrections to the mean field results, these will produce no qualitative alterations in the static limit to the conclusions that follow. They are a must for calculations at finite temperatures and high fields, but those conditions are not met in the experiments of interest here; for example, in the ESR experiment of Ref. 6, the spectrometer frequency was set typically at 9.3 GHz, or about 0.5 K in temperature units, small compared to the characteristic temperature of $\mathrm{UBe}_{13}$ of about 10 K.

While the above model is suggested by the various mean field treatments of the Anderson lattice problem, essentially identical conclusions to those discussed below also follow from a Green's function treatment where the dispersion of the $f$-electron self-energy (apart from the one-body hybridization terms) is ignored. We defer this derivation to the Appendix, since the physical ideas are clearly exposed within the framework of the mean-field theory.

The Hamiltonian of Eq. (10) is easily diagonalized. This simple form results in two hybridized bands, the lower of which has energies $E_{k\alpha}$ given by

$$E_{k\alpha} = 1/2\{(\varepsilon_k + \varepsilon_r) - [(\varepsilon_k - \varepsilon_r)^2 + 4\tilde{V}^2]^{1/2}\} \tag{11}$$

has states given by

$$\alpha^\dagger_{k\sigma} = u_k c^\dagger_{k\sigma} + v_k f^\dagger_{k\sigma} \tag{12}$$

with $u_k = (E_{k\alpha} - \varepsilon_r)/[(E_{k\alpha} - \varepsilon_r)^2 + \tilde{V}^2]^{1/2}$. The density of states of the lower band is

$$\tilde{N}(E_{k\alpha}) = N(\varepsilon(E_{k\alpha}))/u^2_{k\alpha} \; , \tag{13}$$

where $N(\varepsilon(E_{k\alpha}))$ is the bare conduction electron density of states evaluated at the energy $\varepsilon(E_{k\alpha})$ equal to $E_{k\alpha} - \tilde{V}^2/(\varepsilon_r - E_{k\alpha})$.

Now I assume simple exchange coupling of the probe spin to the original conduction and $f$-electron quasiparticles described by Eq. (10). Explicitly, for probe spin $\mathbf{S}_n$, I assume a contact coupling with the ordinary conduction states

$$H^c_{\mathrm{exch}} = -\frac{J_{nc}}{N_s} \sum_{\mathbf{k},\mathbf{k}',\mu,\nu} \mathbf{S}_n \cdot \boldsymbol{\sigma}_{\mu\nu} c^\dagger_{\mathbf{k}\mu} c_{\mathbf{k}'\nu} \; , \tag{14}$$

and a coupling to the $f$ states given by

$$H^f_{\mathrm{exch}} = -\frac{1}{N_s} \sum_{\mathbf{k},\mathbf{k}',\mu,\nu} \mathbf{S}_n \cdot \boldsymbol{\sigma}_{\mu\nu} J_{nf}(\mathbf{k}-\mathbf{k}') f^\dagger_{\mathbf{k}\mu} f_{\mathbf{k}'\nu} \; , \tag{15}$$

where the exchange integral is given in terms of the site basis as

$$J_{nf}(\mathbf{k}-\mathbf{k}') = \sum_{\mathbf{R}(\neq 0)} e^{i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{R}} J_{nf}(\mathbf{R}) \; . \tag{16}$$

The relevant combination of exchange integrals for the low temperature and frequency relaxation of the probe spin in terms of the hybridized quasiparticles is that for intraband exchange within the $\alpha$ band, since there will be a gap for excitation of interband exchange processes at low temperature and frequency. The intraband exchange is given by

$$J_{n\alpha}(\mathbf{k}-\mathbf{k}') = u_k u_{k'} J_{nc} + v_k v_{k'} J_{nf}(\mathbf{k}-\mathbf{k}') \; . \tag{17}$$

In terms of this intraband exchange, the probe relaxation rate in the limit of low-frequency, field, and temperature is given by the formula

$$\lim_{T\to 0}\frac{1}{T_1 T}=\frac{\mu_{\rm eff}^2}{3(g\mu_B)^2}\frac{N_{\rm grd}\pi k_B}{N_s^2\hbar}\sum_{\mathbf{k},\mathbf{k}'}J_{n\alpha}^2(\mathbf{k}-\mathbf{k}')f(E_{k\alpha})[1-f(E_{k'\alpha})]\delta(E_{k\alpha}-E_{k'\alpha})\ . \tag{18}$$

The above expression can be readily manipulated into the form

$$\lim_{T\to 0}\frac{1}{T_1 T}=\frac{\mu_{\rm eff}^2}{3(g\mu_B)^2}\frac{N_{\rm grd}\pi k_B}{\hbar}[N(\varepsilon(0))J_{nc}]^2\left\{\left[1+\left[\frac{\tilde V}{\varepsilon_r}\right]^2\frac{[J_{nf}]_{\rm FS}}{J_{nc}}\right]^2+\left[\frac{\tilde V}{\varepsilon_r}\right]^4\frac{[J_{nf}^2]_{\rm FS}-[J_{nf}]_{\rm FS}^2}{J_{nc}^2}\right\}, \tag{19}$$

where the square bracket Fermi surface (FS) average denotes, e.g.,

$$[J_{nf}]_{\rm FS}=\int\int\frac{d\hat{\mathbf{k}}\,d\hat{\mathbf{k}}'}{(4\pi)^2}J_{nf}(\mathbf{k}-\mathbf{k}')\,|_{E_{k\alpha}=E_{k'\alpha}=0}\ . \tag{20}$$

Here, of course, $\hat{\mathbf{k}}=\mathbf{k}/k$ is the direction of $\mathbf{k}$. I note that the factor $\tilde V/\varepsilon_r$ is approximately $(D/T_0)^{1/2}$, which is precisely the square root of the effective mass ratio.

Equation (19) has two interesting limits. The first case is when $J_{nf}$ tends to zero, so that all relaxation of the probe spin is due to on-site coupling the normal conduction electrons. I assert that this is the limit relevant to impurity based ESR in most instances, and I will return to that point later in the manuscript. This limit is where the many-body cancellation arguments apply since the effective mass explicitly drops from the expression. Very explicitly, on converting sum to integral of Eq. (19) contains the following combination of mass factors in this limit

$$\frac{N(\varepsilon(0))^2}{u_k^4}u_k^4 J_{nc}\ , \tag{21}$$

the factors in the denominator coming from wave-function renormalization modification of the density of states, and the factors in the denominator coming from the renormalization of the interaction between the probe and conduction electron moments. The relaxation rate is different from that in the absence of $f$ electrons only by the factor $[N(\varepsilon(0))/N(0)]^2$. This would not be expected to lead to anything like an order of magnitude difference, and rather than lead to enhancement is more likely to lead to suppression of the relaxation rate due to the fact that $\varepsilon(0)\sim D$ and one expects the density of states near the band edges to smaller than that in the middle. However, any quantitative estimate of the effect is beyond the scope of this paper.

We note the conclusions in the above paragraph are unmodified with respect to the cancellation of the effective mass factors (identified as $1/u_{k\alpha}^2$) when realistic dispersion of the matrix element $\tilde V$ is allowed; a calculation which includes the possible dispersion of the hybridization gives the following result:

$$\lim_{T\to 0}\frac{T_1^0}{T_1}=\left\langle\frac{1}{|\mathbf{v}_\mathbf{k}|}\right\rangle_{\rm FS}^2\Big/\left\langle\frac{1}{|\mathbf{v}_\mathbf{k}^0|}\right\rangle_{\rm FS}^2\ . \tag{22}$$

Here $T_1^0$ is the spin-lattice relaxation in the absence of hybridization, $T_1$ is the relaxation time for $V\neq 0$, the angular brackets imply an angular average over the respective Fermi surfaces, $\mathbf{v}_\mathbf{k}^0$ is the conduction electron velocity for $V=0$, and $\mathbf{v}_\mathbf{k}$ measures the modified velocity and is given by

$$\mathbf{v}_k=\mathbf{v}_\mathbf{k}^0-2\frac{\tilde V_\mathbf{k}}{\varepsilon_r}\frac{\partial\tilde V_\mathbf{k}}{\partial\mathbf{k}}\ . \tag{23}$$

It is understood that $v_k^0$ in the above expression is to be evaluated at $\varepsilon_\mathbf{k}=\varepsilon_0$. Note that depending upon the sign of the $\mathbf{k}$ gradient of the hybridization, the numerator of Eq. (22) could lead either to suppression or enhancement of the on-site relaxation in the presence of the $f$ electrons.

It should be pointed out that in Ref. 8(a), a similar calculation was carried out in this limit where on-site relaxation dominates. The differences from the calculation presented here are (i) the calculation was done for a single rare-earth heavy-electron site (the modifications to the conduction-electron Green's function due to scattering off of the distant site were considered), and (ii) the resonance parameters were not fixed in the self-consistent fashion implicit here, as a physical picture closer to the strongly mixed valent limit was envisioned, with the bare $f$ level approaching the Fermi level.

The second interesting limit for spin-lattice relaxation of the probe moment given by Eq. (19) is when the probe-$f$ exchange obeys the inequality

$$\left|\frac{D[J_{nf}]_{\rm FS}}{T_0}\right|\gg|J_{nc}|\ . \tag{24}$$

In this limit, the $f$-electron spin fluctuations dominate the relaxation of the probe moment, and this is the case for typical NMR probes as I have argued in the first section of the paper and will consider again below in a different context. In fact, the relaxation rate in this limit is little different from the incoherent rate calculated in the previous section. To demonstrate this, I note that explicitly the relaxation rate tends to

$$\lim_{T\to 0}\frac{1}{T_1 T}=\frac{\mu_{\rm eff}^2}{3(g\mu_B)^2}\frac{N_{\rm grd}\pi k_B}{\hbar}\left[\frac{N(\varepsilon(0))\tilde V^2}{\varepsilon_r}\right]^2[J_{nf}^2]_{\rm FS}$$
$$\sim\frac{\mu_{\rm eff}^2}{3(g\mu_B)^2}\frac{\pi k_B}{\hbar}\frac{[J_{nf}^2]_{\rm FS}}{N_{\rm grd}T_0^2}\ . \tag{25}$$

The average of $J_{nf}^2$ is given by

$$[J_{nf}^2]_{\rm FS}=\sum_{\mathbf{R},\mathbf{R}'(\neq 0)}J_{nf}(\mathbf{R})\left[\frac{\sin(k_0|\mathbf{R}-\mathbf{R}'|)}{k_0|\mathbf{R}-\mathbf{R}'|}\right]^2 J_{nf}(\mathbf{R}')$$
$$\approx\sum_{\mathbf{R}(\neq 0)}J_{nf}^2(\mathbf{R})\left[1+O\left[\frac{1}{(k_0a)^2}\right]\right]\ . \tag{26}$$

This result follows since the factor $[\sin(kr)/kr]^2$ acts like a Kronecker $\delta$ function. In the above, $k_0$ is defined as the wave vector corresponding to $\varepsilon(0)$. Thus, the mean-field model reproduces the incoherent picture (sum on sites of the on-site value of $\chi''(\omega)/\omega$) in the parameter regime determined by Eq. (24) to within factors of order unity [Eq. (26) giving only a few percent correction].

The dipolar mechanism which is central to the paper is an obvious example of where this limit implied by (24) applies, and although the above result is derived strictly for exchange interactions it is straightforward to extend the derivation to dipolar coupling. It is clearly the matrix element effect implicit within (25) which prevents the Be moments from having a millionfold enhancement as naively guessed on comparing the specific heats of $UBe_{13}$ and $ThBe_{13}$.

The probability of a weak matrix element in the case of ESR, as alluded to earlier, is, I believe, the most likely reason that the impurity electronic moments are not effective probes of the heavy electron state. To clarify this point, let us assume that the interaction between the impurity probe and the $f$ electrons is indeed of the RKKY form. Then, roughly, if I put the impurity at the origin,

$$J_{nf}(\mathbf{R}) \approx \frac{J_{nc}}{J_{cf}} I_{\mathrm{RKKY}}(\mathbf{R}) , \tag{27}$$

where $J_{cf}$ is the bare conduction-electron–$f$-electron exchange interaction, and $I_{\mathrm{RKKY}}$ is the RKKY interaction which would have existed between a heavy electron atom at the probe site and the $f$ moment at $\mathbf{R}$. One expects this quantity to scale roughly with the paramagnetic transition temperature $T_p$. Hence, we can estimate the maximal $f$-electron enhancement of the ESR relaxation rate as

$$\left. \left[ \frac{\tilde{V}}{\varepsilon_r} \right]^4 \frac{[J_{nf}^2]_{\mathrm{FS}}}{J_{nc}^2} \right|_{\max} \approx \frac{1}{q_E} \left[ \frac{D}{J_{cf}} \right]^2 \left[ \frac{T_p}{T_0} \right]^2 , \tag{28}$$

where $q_E$ is the nearest $f$-shell coordination about the impurity site. While one can generally expect $D/J_{cf} \sim 20$, in those compounds like $UBe_{13}$ and $CeBe_{13}$ which do not magnetically order, it is probably the case that $T_P/T_0 < 0.1$, say, so that it is unlikely that the maximal $f$-electron enhancement will exceed unity. "Best case" estimates of (28) for $UBe_{13}$ and $CeBe_{13}$ gives upper bounds of 10 and 0.025, respectively, for the maximal enhancement, assuming the simplest free electron form for the RKKY interaction, a conduction bandwidth of 3 eV, $k_F \approx 1$ Å$^{-1}$, and $J_{cf}/D \approx 0.05$. Clearly, a reliable calculation awaits a reliable calculation of the band structure of these materials taking the near integral occupancy of the $f$ electrons into account. This is a project for the future.

It should be noted that Eq. (28) suggests that ESR on those materials which magnetically order might well provide a more fruitful probe of the heavy-electron state, since the rough criterion for magnetic order is that $T_P/T_0 > 1$.[48,51] Of course, in that limit it would be very important to take account of the intersite correlations in evaluating Eq. (18), which goes beyond the scope of this paper. Some specific suggestions would be $CeAl_2$, $CeB_6$, $U_2Zn_{17}$, and $UCd_{11}$. While the antiferromagnetic fluctuations above the magnetic transitions in these materials might complicate the interpretation, it is possible to suppress the transition by doping (La or Th) on the (Ce or U) sublattice. For the first two materials, this could lead to a rather clean probe of very heavy electrons since the characteristic temperature is of order 1 K for all concentrations of Ce atoms.

Finally, I contrast Eq. (28) with the maximal $f$-electron enhancement for a heavy nucleus probe moment. In that case, as discussed in the preceeding section, I cannot expect dipolar coupling to be relevant and RKKY again must dominate. The expression corresponding to (28) is

$$\left. \left[ \frac{\tilde{V}}{\varepsilon_r} \right]^4 \frac{[J_{nf}^2]_{\mathrm{FS}}}{J_{nc}^2} \right|_{\max} \approx \frac{q_N}{q_E^2} \left[ \frac{D}{J_c} \right]^2 \left[ \frac{T_P}{T_0} \right]^2 \left[ \frac{a_E}{a_N} \right]^6 , \tag{29}$$

where $q_N$ is the nearest $f$-shell coordination about the nucleus at a distance $a_N$, and $a_E$ is the corresponding distance in the ESR case. The "best case" analysis corresponding to that below Eq. (28) yields values of 100 and 0.25, respectively, for the maximal enhancements of $UBe_{13}$ and $CeBe_{13}$, compared with the observed enhancements of order 1000 and 10.[4,52]

While the exact power law of the distance ratio in Eq. (29) should not be taken too seriously, since it will, in general, depend upon direction and the actual shape of the normal conduction electron Fermi surface, the point that the simple distance dependence makes such an enormous difference in the observed relaxation behavior is clearly demonstrated. The NMR probe sites, as witnessed in Table II, sit typically at about 3 Å from the heavy electron sites, while $f$-$f$ spacings are usually 4–5 Å, the latter holding for $UBe_{13}$. The ratio of $(3/5)^6$ is about $\frac{1}{30}$. One might hope to see significant enhancement in ESR on materials where the $f$-$f$ spacing is unusually small;[12] unfortunately, this will usually either correlate with lower specific heat coefficients due to increased hybridization or in more anisotropic cases, magnetic order due to enhanced nearest-neighbor exchange [as happens in $CeRh_3B_2$ (Ref. 53)].

## SUMMARY

In summary, I have shown that the dipolar coupling of $4f$ or $5f$ spin fluctuations to $^9$Be nuclear moments is sufficient to understand the enhanced spin lattice relaxation rates of $UBe_{13}$ and $CeBe_{13}$. This mechanism cannot, however, play any significant role in the relaxation of heavier nuclei (such as $^{119}$Sn in $CeSn_3$) which have already strong intrinsic relaxation due to on-site coupling to the conduction electrons. Neither can the dipole mechanism make a significant contribution to the relaxation of say, Er moments in $UBe_{13}$, where only RKKY coupling with the heavy electrons and on-site exchange with the normal electrons are feasibly strong. The cancellation arguments put forth to understand the lack of relaxation enhancement in ESR apply only to the contact coupling to ordinary conduction electron states and clearly are less relevant in the NMR case, for light nuclei at least. Should the on-site couplings ($J_{nc}$, schematically) be very strong, then the relaxation rate will be subject to the cancellation arguments, apart from a modification of the energy at which the bare density of states is evaluated. Rather than being evaluated at the zero hybridization Fermi level, it is evaluated near the band edge which should tend to lead to suppression since for most bands, the density of states is higher in the middle of the band. In addition, if the hybridization is dispersive, while the mass effects still cancel, the rate may be enhanced or suppressed depending upon the **k** gradient of the hybridization at the Fermi level. Any direct interaction between $4f$ or $5f$ moments and probe spins can in principle probe the enhanced, heavy electron density of states to the extent that the range dependence (nominally $\sim 1/R^6$ for both dipolar and RKKY interactions) of the matrix elements is not too great. This range dependence is clearly of great relevance for both ESR and NMR where there is typically a large-probe–heavy-electron site separation in the former case of about 4–5 Å and usually a separation of only about 3 Å in the latter. The lack of success so far in using ESR to probe the heavy-electron state might be turned around in those heavy electron compounds which magnetically order, since the maximal enhancement due to the $f$ electrons is roughly proportional to $(D/J_{cf})^2(T_P/T_0)^2$, where $T_P$ is the paramagnetic transition temperature, $D$ is the conduction bandwidth, and $J_{cf}$ is the conduction-electron exchange interaction. Specific candidates for such experiments are $CeAl_2$, $CeB_6$, $CePb_3$, $U_2Zn_{17}$, and $UCd_{11}$.

*Note added.* In the course of submitting this article for publication, two more facts have been brought to light. First, the Knight shift data for single crystal $UBe_{13}$ have been analyzed by MacLaughlin and Clark and it is clear that the isotropic component is of roughly the same size as the anisotropic component which may be presumed due to dipolar coupling. This means that some of the relaxation is due to the same mechanism responsible for the isotropic shift, which may well be RKKY interactions between the probe nuclei and the $f$ electrons.[54] Second, ESR measurements on impurities in $UPt_3$ have yielded similar results as for the corresponding impurities in $UBe_{13}$.[55] Finally, I have recently received a copy of work prior to publication from Simanek and Sasahara[56] which arrives at similar conclusions concerning the relevance of cancellation effects in the limit of strong on-site coupling via a Green's function formalism closely related to that of the Appendix. I thank these authors for sharing their results prior to publication.

## ACKNOWLEDGMENTS

I have benefitted greatly from discussion with L. J. Sham, F. Gandra, S. Schultz, D. MacLaughlin, W. G. Clark, J. W. Wilkins, N. E. Bickers, R. Cotts, M. Dubson, J. Markert, A. Ruckenstein, S. Oseroff, P. Fulde, A. J. Millis, and S. D. Mahanti. I have also benefitted from a correspondence with H. Schäffer, M. Schlott, and J. Elschner. I thank A. J. Millis and P. A. Lee for sending me a copy of their work prior to publication. This research was supported by National Science Foundation Grant No. DMR85-14195. I am grateful for the hospitality of the Institute of Theoretical Physics where some of this work was performed and for partial support there through National Science Foundation Grant No. PHY82-17853 supplemented by funds from the National Aeronautics and Space Administration.

## APPENDIX: GREEN'S FUNCTION DERIVATION OF LATTICE RESULTS

In this appendix, I derive a formula which agrees in essential detail with the mean-field theory result of Eq. (18) by a Green's function approach where it is assumed that the interaction piece of the $f$-electron self-energy is dispersionless. Thus the derivation is expected to be relevant to the extent that intersite correlations between the $f$-electron moments are small; i.e., it is most applicable to the heavy electron materials which do not magnetically order.

The model Hamiltonian is the full Anderson lattice Hamiltonian, given by

$$H=\sum_{k,\sigma}[\varepsilon_k c_{k\sigma}^{\dagger}c_{k\sigma}+\varepsilon_f f_{k\sigma}^{\dagger}f_{k\sigma}+V(c_{k\sigma}^{\dagger}f_{k\sigma}+\mathrm{H.c.})]+U\sum_{R,\sigma(\neq\sigma')}n_{fR\sigma}n_{fR\sigma'}\ . \tag{A1}$$

We assume $U \gg |\varepsilon_f| \gg \Gamma=\pi N(0)V^2$ to put us in the asymmetric local moment regime.

As is well known, the electron Green's functions for spin index $\sigma$ can be written formally as a matrix with "isotropic" indices $\alpha$ running over $c$, $f$, and entries given by

$$G_{cc\sigma}(\mathbf{k},\omega,T)=[\omega-\varepsilon_k-V^2/(\omega-\varepsilon_f-\Sigma(\mathbf{k},\omega,T))]^{-1}\ , \tag{A2a}$$

$$G_{ff\sigma}(\mathbf{k},\omega,T)=[\omega-\varepsilon_f-\Sigma(\mathbf{k},\omega,T)-V^2/(\omega-\varepsilon_k)]^{-1}\ , \tag{A2b}$$

and

$$G_{fc\sigma}(\mathbf{k},\omega,T)=VG_{cc}^{0}(\mathbf{k},\omega)G_{ff}(\mathbf{k},\omega,T)\ . \tag{A2c}$$

Several authors have utilized these forms.[57–59] In the above, $G_{cc}^{0}(\mathbf{k},\omega)=[\omega-\varepsilon_k]^{-1}$ is the noninteracting conduction-electron Green's function, and $\Sigma(\mathbf{k},\omega,T)$ is that part of the $f$-electron self-energy arising from $f$-$f$ interactions (as opposed to the one body self-energy term arising from mixing). As such, the imaginary part of $\Sigma$ obeys standard Fermi-liquid phase-space arguments[60] and thus vanishes quadratically in $\omega$ at zero temperature.

The important point about $\Sigma(\mathbf{k},\omega,T)$, as has been noted elsewhere, is that for large $U$, one expects the local interactions to dominate and thus for this self-energy to be rather dispersionless.[61] Dispersion becomes relevant to the extent that intersite interactions (of necessity mediated by conduction electrons and thus loosely identified as RKKY interactions) are strong. In the heavy-electron superconductors, valence fluctuation materials, and heavy-electron materials (which show no order of any sort, it is apparently the case that this situation holds at least as a zeroth-order approximation coming in from the high-temperature side.[62]

With this assumption about the structure of $\Sigma$, I can make connection with the parameters of the mean field resonant level lattice. I define $\varepsilon_r$ as

$$\varepsilon_r=\frac{\varepsilon_f+\Sigma(\omega=0,T=0)}{1-\left.\dfrac{\partial\Sigma}{\partial\omega}\right|_{\omega,T=0}} \tag{A3}$$

and assume $\varepsilon_r \sim T_0$. I define $\tilde{V}$ via

$$\tilde{V}^2=V^2\Big/\left[1-\left.\frac{\partial\Sigma}{\partial\omega}\right|_{\omega,T=0}\right] \tag{A4}$$

and assume $\tilde{V}^2 \sim DT_0$. As before, the parameter $\varepsilon(0)$ is $\tilde{V}^2/\varepsilon_r$. Yosida and Yamada have noted that there will be a contribution to the wave-function renormalization from equal spin interactions (ruled out in the totally dispersionless self-energy model) to the extent that the susceptibility of the $f$ electrons possesses momentum dependence.[58]

It is a straightforward exercise in finite-temperature diagrammatic perturbation theory to evaluate the relaxation rate of the magnetic probe. The basic result which has a well-known structure[17] (generalized here to include the "isotopic" indices) is

$$\lim_{T\to 0}\frac{1}{T_1T}=\frac{k_B}{\hbar N_s(g\mu_B)^2}\sum_{q,\alpha,\beta}J^0_{n\alpha}(\mathbf{q})J^0_{n\beta}(\mathbf{q})\lim_{\omega,T\to 0}\frac{\chi''_{\alpha\alpha\beta\beta}(\mathbf{q},\omega,T)}{\omega} . \tag{A5}$$

The relevant diagrams for $\chi''$ are shown in Fig. 1 (the $\omega$ differentiation being taken account of in that figure). I shall elucidate the differences between $J^0(\mathbf{q})$ and $J(\mathbf{q})$ [Eq. (14)] below.

In evaluating the low-frequency dynamic susceptibility ratios which appear in (A5), I follow Shiba[23] in noting that it is the singular part of the Green's functions which appear between the vertices in Fig. 1 which will contribute. One can follow standard lines of analysis[63] to show how the product separates into a regular and singular part for low frequency $\omega$ according to

$$G_{\alpha\beta\sigma}(p+q)G_{\beta\alpha\sigma}=[G_{\alpha\beta\sigma}(p+q)G_{\beta\alpha\sigma}(p)]_{\text{reg}}+2i\omega\delta(p_0)\rho_{\alpha\beta\sigma}(p+q)\rho_{\beta\alpha\sigma}(p) . \tag{A6}$$

In the above, $p=(p_0,\mathbf{p})$ and $q=(\omega,\mathbf{q})$ are four-vectors and the spectral functions $\rho_{\alpha\beta\sigma}(p)$ are defined by

$$\rho_{\alpha\beta\sigma}(p)=-\operatorname{Im}G_{\alpha\beta\sigma}(p_0+i\eta,\mathbf{p}),\eta\to 0^+ . \tag{A7}$$

The differentiation with respect to $\omega$ breaks a line in each diagram of Fig. 1, and therefore, one has vertex corrections at each vertex. The vertex function $\Lambda_{\alpha\sigma}(p+q,p)$ is just $\sigma$ for $\alpha=c$, and is given, for $\alpha=f$, by

$$\begin{aligned}\Lambda_{f\sigma}(p+q,p)&=\sigma+\sum_{\sigma'}\int dp' I_{f\sigma\sigma'}(p,p',q)G_{ff\sigma'}(p'+q)G_{ff\sigma'}(p')\Lambda_{f\sigma'}(p'+q,p')\\&=\sigma+\sum_{\sigma'}\sigma'\int dp'\Gamma_{ff\sigma'}G_{ff\sigma'}(p'+q)G_{ff\sigma'}(p') ,\end{aligned} \tag{A8}$$

where $I_{ff\sigma}$ and $\Gamma_{ff\sigma}$, are, respectively, the irreducible and reducible four-point interaction functions of the $f$ electrons. For $q\to 0$ with $\omega/|\mathbf{q}|\to 0$, (A8) is solved by

$$\begin{aligned}\Lambda_{f\sigma}&=\sigma\left[1-\left.\frac{\partial\Sigma(p)}{\partial h_{f\sigma}}\right|_0+\left.\frac{\partial\Sigma(p)}{\partial h_{f-\sigma}}\right|_0\right]+O(T_0/D)\\&=\sigma\lambda_f(\mathbf{q}=0) ,\end{aligned} \tag{A9}$$

where $h_{f\sigma}$ is a field which couples only to $f$ spin $\sigma$ (see Ref. 58). In terms of this vertex, the static susceptibility is given by

$$\chi_{ff}(0)=\frac{\mu^2_{\text{eff}}}{3}\frac{\lambda_f(0)}{1-\left.\dfrac{\partial\Sigma}{\partial\omega}\right|_0}N_{\text{grd}}\left[\frac{N(\varepsilon(0))\tilde{V}^2}{\varepsilon_r^2}\right] . \tag{A10}$$

Inserting (A6) and (A9) into (A5) and defining the reduced vertex function $\zeta_f(\mathbf{q})$ by

$$\zeta_f(\mathbf{q})=\frac{\lambda_f(\mathbf{q})}{\lambda_f(0)}=\frac{\Lambda_{f\sigma}(0,\mathbf{p},0,\mathbf{p}+\mathbf{q})}{\sigma\lambda_f(0)} , \tag{A11}$$

I find that ($\mathbf{q}=\mathbf{k}-\mathbf{k}'$)

$$\begin{aligned}\lim_{T\to 0}\left[\frac{1}{T_1T}\right]&=\left[\frac{\mu^2_{\text{eff}}}{3(g\mu_B)^2}\right]\frac{\pi k_B N_{\text{grd}}}{\hbar N_s^2}\\&\quad\times\sum_{k,k',\alpha,\beta}J_{n\alpha}(\mathbf{q})J_{n\beta}(\mathbf{q})\lambda_\alpha(0)\\&\quad\times\rho_{\alpha\beta\sigma}(\mathbf{k},0)\rho_{\beta\alpha\sigma}(\mathbf{k}',0)\lambda_\beta(0) ,\end{aligned} \tag{A12}$$

where $\lambda_c(0)$ is identically one and I define

$$J_{nf}(\mathbf{q})=\zeta_f(\mathbf{q})J^0_{nf}(\mathbf{q}) . \tag{A13}$$

Note that (A13) is the desired relation between the bare interaction and the quasiparticle interaction given by Eq. (14).

Note that the diagram of Fig. 1(d) which represents a possible nontrivial vertex correction to the *cccc* term in Eq. (A5) can in fact be incorporated into the *ffff* term with a suitable redefintion of $J_{nf}$. Hence, I shall assume that all such conduction-electron-mediated interactions between the probe and $f$ electrons are included in $J_{nf}$ at

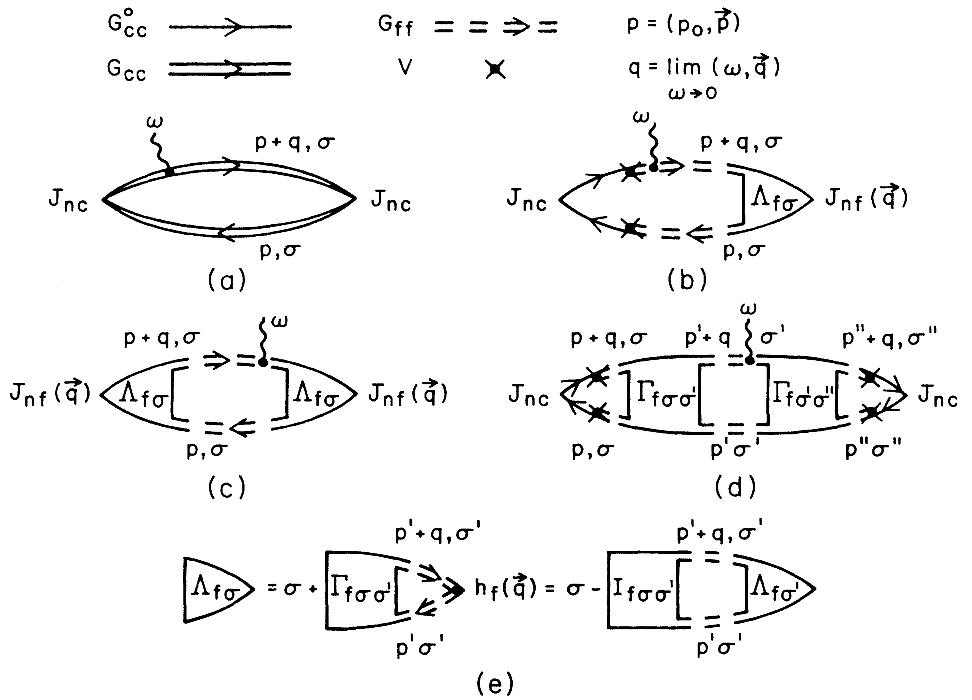

FIG. 1. Relevant diagrams for Green's function of $1/T_1T$. The correspondence of the various lines to the propagators of Eqs. (A2a)–(A2c) is shown at top. The wavy line with an $\omega$ at its tip corresponds to differentiation by $\omega$ as must happen according to (A5). Figure 1(a) corresponds to the "normal" on-site relaxation, Fig. 1(b) corresponds to mixed relaxation which exists in the presence of hybridization due to the "off-diagonal" propagators of Eq. (A2c), and Fig. 1(c) corresponds to the magnetic probe relaxation induced by the spin fluctuations of the $f$ electrons. Note that while Fig. 1(d) shows a potential nontrivial vertex correction to Fig. 1(a), it is more properly incorporated into a redefinition of the bare $J_{nf}$ as discussed in the text. The relationship between the three point vertex $\Lambda_{f\sigma}$ and the interaction functions $I_{f\sigma\sigma'}$ (irreducible four-point interaction) and $\Gamma_{f\sigma\sigma'}$ (reducible four-point interaction) is illustrated in Fig. 1(e).

the outset and in that way avoid double counting diagrams.

Since the spectral functions of (A12) are constrained to the Fermi surface, then the Fermi liquid phase space considerations enter, and the spectral functions defined by (A7) tend to δ functions. Explicitly,

$$\rho_{cc}(\mathbf{k},\omega=0,T=0)=\pi\delta(\varepsilon_k-\tilde{V}^2/\varepsilon_r)\;, \tag{A14a}$$

$$\rho_{ff}(\mathbf{k},\omega=0,T=0)=\frac{\pi\delta(\varepsilon_r-\tilde{V}^2/\varepsilon_k)}{1-\left.\dfrac{\partial\Sigma}{\partial\omega}\right|_{\omega,T=0}}\;, \tag{A14b}$$

$$\rho_{cf}(\mathbf{k},\omega=0,T=0)=\frac{-V}{\varepsilon_k}\rho_{ff}(\mathbf{k},\omega=0,T=0)\;. \tag{A14c}$$

Substitution of (A14a)–(A14c) into (A12) yields the desired expression for the relaxation rate which is

$$\lim_{T\to 0}\frac{1}{T_1T}=\frac{\mu_{\text{eff}}^2}{3(g\mu_B)^2}\,\frac{\pi k_B}{2\hbar}[N(\varepsilon(0))J_{nc}]^2\left\{\left[1+\left[\frac{\tilde{V}}{\varepsilon_r}\right]^2 R\,\frac{[J_{nf}]_{\text{FS}}}{J_{nc}}\right]^2+\left[\frac{\tilde{V}}{\varepsilon_r}\right]^4 R^2\left[\frac{[J_{nf}^2]_{\text{FS}}-[J_{nf}]_{\text{FS}}^2}{J_{nc}^2}\right]\right\}, \tag{A15}$$

where the "Wilson ratio" $R$ is given by

$$R=\frac{\lambda_f(0)}{1-\left.\dfrac{\partial\Sigma}{\partial\omega}\right|_0}\;. \tag{A16}$$

The only difference from the mean-field result is the appearance of the factor $R$. Since $R$ is expected to be of order unity and since the mean-field theory is apparently capable of including $R$ once Gaussian fluctuations are accounted for,[64,65] then I conclude that there is no essential discrepancy between the Green's function approach and the mean-field approach.

Of course, in practice, there can be an important renormalization of the interactions through (A13). For a dispersionless self-energy one might expect the reduced vertex $\zeta_f(\mathbf{q})$ to be constant over the Fermi surface. To gauge this notion, for simplicity I assume a spherical Fermi surface and expand in spherical harmonics

$$\zeta_f^2(\mathbf{k}-\mathbf{k}')=\sum_{l=0}^{\infty}(2l+1)[\zeta_f^2]_l P_l(\hat{\mathbf{k}}\cdot\hat{\mathbf{k}}') \ . \tag{A17}$$

Peforming the same sort of analysis that leads to (24), I find that [$j_l(x)$ is the $l$th spherical Bessel function]

$$|J_{nf}^2]_{\mathrm{FS}}=\sum_{l=0}^{\infty}(-1)^l(2l+1)[\zeta_f^2]_l \times \sum_{\substack{R,R' \\ (R\neq R')}} J_{nf}^0(\mathbf{R})j_l^2(k_0|\mathbf{R}-\mathbf{R}'|)J_{nf}^0(\mathbf{R}') \ . \tag{A18}$$

Truncating at $l=0$ reproduces (24); keeping $l>0$ will result in nonlocal corrections as $j_l^2(k_0|\mathbf{R}-\mathbf{R}'|)$ acts as a Kronecker $\delta$ function which peaks near $\mathbf{R}=\mathbf{R}'+\boldsymbol{\delta}_l$, where $|\boldsymbol{\delta}_l|>0$. Thus the coefficients in the expansion (A17) provide some crude measure of intersite correlation for $l>0$, to the extent that they do not vanish.

In closing, I note that in the limit implied by (22), the spin lattice relaxation obeys a lattice analogue of the Shiba relation to the extent that the self-energy is dispersionless, since (A16) can be rewritten as

$$\lim_{T\to 0}\frac{1}{T_1T}=\frac{k_B}{\hbar}\frac{1}{(g\mu_B)^2}\left\{\frac{\pi}{N_{\mathrm{grd}}}\frac{3}{\mu_{\mathrm{eff}}^2}[\chi(0)]^2\right\}[J_{nf}^2]_{\mathrm{FS}} \ . \tag{A19}$$

This should be compared with (4), noting that all other factors just give the hyperfine field constants, albeit with a possible nontrivial renormalization of the bare hyperfine couplings.

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