On the ratio T_2/T_1 for non-Ohmic spectral densities

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(Received 29 December 1993; accepted 22 March 1994)

There has been some recent interest[1-8] in the ratio T_2/T_1, where T_2 and T_1 are the phase and population relaxation times, respectively, for a two-level system (TLS). Budimir and Skinner[1] showed that when the TLS is strongly coupled to a stochastic bath, under certain circumstances one finds the unusual result that $T_2>2T_1$. The validity of this result was later extended to finite temperatures by considering a TLS coupled to a quantum-mechanical harmonic bath.[3,9] In this calculation the spectral density of the bath was Ohmic, that is, it was proportional to frequency in the low-frequency limit. Given that for other types of spin-boson Hamiltonians (e.g., the tunneling problem[10,11] and the pure dephasing problem[12]) Ohmic and non-Ohmic spectral densities give qualitatively different results, it is natural to wonder whether the possibility that $T_2>2T_1$ is specific to the Ohmic model, or is, in fact, more generally valid.

To address this question, in this Note we consider the super-Ohmic spectral density used recently by Suárez *et al.*[13]

$$\Gamma_1(\omega)=A(\omega/\omega_c)^3 e^{-\omega/\omega_c} \quad (\omega>0), \tag{1}$$

which has the same low-frequency behavior as the Debye model together with the standard deformation potential approximation. (Herein we follow exactly the definitions and notation of Ref. 3.) For the "complex" coupling model $[\Gamma_2(\omega)=0]$, it was found that[3]

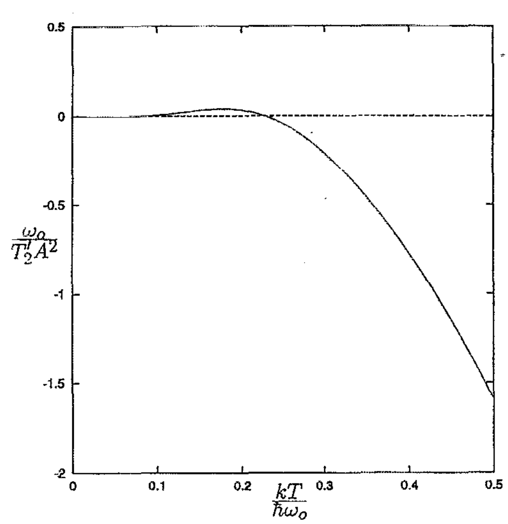

FIG. 1. $\omega_o/T_2'A^2$ vs $kT/\hbar\omega_o$ for $\omega_c/\omega_o=0.4$.

$$\frac{1}{T_2'}=\frac{4\delta^4}{\pi} P\int_{-\infty}^{\infty} \frac{d\omega\ \omega}{\omega^2-\omega_o^2} \times \frac{\partial}{\partial\omega}\{\Gamma_1(|\omega|)^2 n(|\omega|)[n(|\omega|)+1]\}+O(\delta^6), \tag{2}$$

where $1/T_2'\equiv 1/T_2-1/2T_1$, δ is the dimensionless coupling constant, P is the Cauchy principal value, ω_o is the TLS frequency, and $n(\omega)=(e^{\hbar\omega/kT}-1)^{-1}$. In the high-temperature limit we can write (setting $\delta=1$) $1/T_2'=(AkT/\hbar)^2\omega_o^{-3} f(\omega_c/\omega_o)$, and the dimensionless function f can be evaluated numerically. We find that for $0.281<\omega_c/\omega_o<1.006$, $f(\omega_c/\omega_o)<0$, which means that $1/T_2'<0$, and that $T_2>2T_1$. Choosing now the value of $\omega_c/\omega_o=0.4$, we can calculate the temperature dependence of $1/T_2'$, which is shown in Fig. 1. We find that for $kT/\hbar\omega_o>0.228$, $T_2>2T_1$.

Thus, we see that, similar to the Ohmic case, for a reasonably wide range of parameters $T_2>2T_1$. This shows that the breakdown of the usual inequality $T_2\leq 2T_1$ is not simply a peculiarity of the Ohmic model, but is indeed a more general result.

Noted added in proof. We thank Philip Pechukas for sending us the fascinating preprint entitled "Reduced dynamics need not be completely positive."

We are grateful for support from the National Science Foundation (Grant No. CHE92-19474).

[1] J. Budimir and J. L. Skinner, J. Stat. Phys. **49**, 1029 (1987).
[2] H. M. Sevian and J. L. Skinner, J. Chem. Phys. **91**, 1775 (1989).
[3] B. B. Laird and J. L. Skinner, J. Chem. Phys. **94**, 4405 (1991).
[4] T.-M. Chang and J. L. Skinner, Physica A **193**, 483 (1993).
[5] P. Reineker, B. Kaiser, and A. M. Jayannavar, Phys. Rev. A **39**, 1469 (1989).
[6] H. Risken, L. Schoendorff, and K. Vogel, Phys. Rev. A **42**, 4562 (1990).
[7] D. D. Traficante, Concepts Magn. Reson. **3**, 171 (1991).
[8] L. Andreozzi, C. Donati, M. Giordano, and D. Leporini, Phys. Rev. A **46**, 6222 (1992).
[9] B. B. Laird, J. Budimir, and J. L. Skinner, J. Chem. Phys. **94**, 4391 (1991).
[10] A. J. Leggett, S. Chakravarty, A. T. Dorsey, M. P. A. Fisher, A. Garg, and W. Zwerger, Rev. Mod. Phys. **59**, 1 (1987).
[11] R. Silbey and R. A. Harris, J. Phys. Chem. **93**, 7062 (1989).
[12] J. L. Skinner and D. Hsu, J. Phys. Chem. **90**, 4931 (1986).
[13] A. Suárez, R. Silbey, and I. Oppenheim, J. Chem. Phys. **97**, 5101 (1992).

0021-9606/94/101(1)/852/1/$6.00