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Results on the energy-relaxation rates of dense two-temperature aluminum, carbon, and silicon plasmas close to liquid-metal conditions

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We present results for the electron-ion energy relaxation coupling constants $g_{ei}(T_e, T_i, \kappa)$ for aluminum, carbon, and silicon plasmas at several electron and ion temperatures T_i, T_e of experimental interest. The calculations use the Fermi golden rule and the Landau-Spitzer model valid at weak electron-ion coupling, as well as the coupled-mode approach suitable for strong coupling. A physically motivated simple derivation of the coupled-mode energy relaxation formula for two-component charged fluids is presented. While the commonly used weak-coupling theories predict relaxation constants relatively independent of the ion temperature, the strong-coupling theory predicts energy relaxation constants that become smaller by an order of magnitude as the ion temperature is lowered.

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I. INTRODUCTION

Electron-ion systems having a ''two-temperature'' distribution are an important class of nonequilibrium systems that can be created using shock waves, intense-laser pulses, or injection of strongly biased electrons into gated microstructures used in electronic devices. The two-temperature system is a result of the different time scales $\tau_{ii} > \tau_{ee}$ for ion-ion $(i\text{-}i)$, and electron-electron $(e\text{-}e)$ relaxation processes. If the system is created by ultrafast laser pulses that deposit energy in the electron subsystem, rapid electron-electron equilibration produces a local electron temperature T_e, while the ions remain at essentially the initial temperature T_i. If shock waves are used, the ion subsystem heats up and slowly exchanges energy to the colder electrons. Equivalent thermalization problems are encountered in astrophysical and planetary research.

For two weakly coupled subsystems, transition rates can be calculated simply, using the Fermi golden rule (FGR) that evaluates the rate as a product of the initial and final densities of states and the square of the matrix element coupling the two systems. The FGR result is a second-order perturbation expansion in the electron-ion interaction U_{ie}. The FGR is not applicable to the strongly interacting case. The problem is still tractable if the ''coupled-modes'' resulting from the strong coupling could be compactly identified. The electron-screened ion excitations in liquid metals, and in dense plasmas, manifest themselves as ion-acoustic modes that are in fact such ''coupled modes.'' As such, we presented a theory of energy relaxation in interacting plasmas using the nonequilibrium method of Keldysh, Martin, and Schwinger, as it is capable of handling the coupled-mode situation in a rigorous manner [1].

In this Rapid Communication we revisit the theory of energy relaxation (ER) and present a more physical analysis of the coupled-mode problem. We make calculations for C, Al, and Si, and examine the ER rates predicted by various theoretical formulations of increasing generality. Unlike other calculations that use point-charge models for ions and other simplifications, we use first-principles pseudopotentials, realistic frequency, and momentum-dependent screening functions, etc., as required by the detailed theory. Our previous numerical results (for Al with the ions held at the melting temperature) suggested that the ER is significantly smaller than the prediction from the usual Landau-Spitzer (LS) formula [2]. Our calculations confirm that (i) the FGR results are smaller than the Landau-Spitzer estimates by about a factor of 2 to 3 for low temperatures; (ii) when coupled modes are included, the ER for aluminum plasmas near the melting point is confirmed to be smaller by an order of magnitude. The calculations for C and Si systems show that the results for aluminum are quite typical. They are of *topical interest* since several experimental studies point to significantly lower ER rates than those predicted by the traditional calculations [3].

II. THEORY

In order to compare and contrast the available theoretical models, we consider the Landau-Spitzer (LS) model, a degeneracy-corrected Landau-Spitzer (DLS) model, the Fermi golden rule result (FGR), a simplified form derived from there, and finally, the coupled-mode formula (CM). The FGR and CM use realistic pseuopotentials and screening functions. The CM is the most general form considered here. Since many variants of LS and DLS are used in the literature, with different, somewhat *ad hoc* ways of incorporating the ion temperature, electron degeneracy, the form of the collision frequency, etc., we will explicitly list the exact forms used by us. The derivation of the CM formula will be given in the Appendix. Thus, the following formulas define the theoretical models used here.

We have set $e = \hbar = m_e = 1$, although the electron mass m_e will sometimes be displayed for clarity. The ER rate per unit volume (containing $\bar{n}$ electrons and $\bar{\rho}$ ions of charge Z per unit volume) is defined by $R = dE_e/dt$, where E_e is the energy in the electron subsystem at time t.

The *temperature*-relaxation rate can be calculated using a mapping of T_e to an equivalent *classical-fluid* temperature

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T_{cf} (see Ref. [4]). Then the E_e is related to a classical kinetic energy $K=(3/2)\overline{n}T_{cf}$, with $T_{cf}=(T_e^2+T_q^2)^{1/2}$, and T_q is a quantum correction given by

$$T_q/E_F=1/(a+b\sqrt{r_s}+cr_s). \tag{1}$$

The Fermi energy E_F is $1/(2\alpha^2 r_s^2)$, where $\alpha=(4/9\pi)^{1/3}$ and r_s is the electron-sphere radius such that $4\pi r_s^3=\overline{n}^{-1}$. The constants appearing in Eq. (1) are $a=1.594$, $b=-0.3160$, and $c=0.0240$.

The main temperature dependence in the ER rate can be factored out by defining the ER-coupling constant $g_{ER}(T_e,T_i,\kappa)$, given by $g_{ER}=R/(T_e-T_i)$. It is expressed in $\mathrm{W\,K^{-1}\,m^{-3}}$ of the material.

A. Landau-Spitzer formula

The Landau-Spitzer (LS) formula for the energy loss rate (ELR) can be written as

$$R_{LS}=\frac{\omega_{ip}^2\omega_{ep}^2(T_e-T_i)\log(\Lambda)}{(2\pi T_e/m_e+2\pi T_i/M_i)^{3/2}}. \tag{2}$$

This is simply the Rutherford formula for Coulomb scattering of particles with Maxwellian velocity distributions at T_e and T_i; for example, $(m_e/2\pi T_e)^{3/2}$ is the normalization constant of the Maxwellian distribution for electrons. The plasma frequencies $\omega_{\tau p}$, where $\tau=e,i$ characterize each subsystem. In our numerical work the Coulomb logarithm $\log(\Lambda)$ is calculated as in Lee and More [5].

B. Degeneracy corrected Landau-Spitzer formula

The Maxwellian distribution for electrons can be replaced by a Fermi distribution to correct for degeneracies. Such a degeneracy-corrected Landau-Spitzer (DLS) form is given by

$$R_{DLS}=R_{LS}/[\sqrt{\pi}(1+e^{-\mu_e/T_e})F_{1/2}(\mu_e/T_e)]. \tag{3}$$

Here μ_e is the electron chemical potential and $F_{1/2}(x)$ is the Fermi integral. This form, with T_i set to zero, has been discussed by Brysk [6]. When $T_i=0$, the resulting collision frequency is consistent with that given by Lee and More in their resistivity calculation.

C. The Fermi golden rule formula (FGR)

In strongly coupled systems, the excitations involve single-particle and collective modes. The energy transfer from the modes of hotter systems to the modes of the cooler system is given by the FGR if the coupling between the systems, denoted by $U_{ie}(q)$, is weak, even if the *intra*system coupling is strong. The two subsystems are characterized by their spectral functions for density excitations, $A_\tau(\omega,q,T_\tau)$. These are evaluated from the imaginary parts of *one-component-fluid* type response functions,

$$A_\tau(q,\omega,T_\tau)=-2\,\mathrm{Im}\,\chi_{\tau\tau}(\mathbf{q},\omega,T_\tau), \tag{4}$$

$$\chi_{\tau\tau}(\omega,\mathbf{q},T_\tau)=\chi^0_{\tau\tau}/\{1-V_{\tau\tau}(q)[1-\mathcal{G}_{\tau\tau}(q)]\chi^0_{\tau\tau}\}. \tag{5}$$

In the above, the zeroth-order response function is denoted by $\chi^0_{\tau\tau}$, where the ω,q,T dependence is suppressed for brevity. This is the usual Lindhard function for electrons, and the plasma dispersion function for ions. More details of the local-field correction $\mathcal{G}_{\tau\tau}$, etc., are given in Refs. [1,4].

The electron-ion interaction is given as a weak pseudopotential having the form $U_{ie}(q)=-Z_iV_qM_q$, where V_q is the Coulomb potential $4\pi/q^2$, and M_q is the matrix element. The appropriate pseudopotentials, derived from first-principles calculations, are given in Ref. [1].

The FGR result, cast into a convenient form, is given by [1]

$$R_{FGR}=\Sigma_{\mathbf{q},\omega}|U_{ie}(q)|^2\omega A^e(q,\omega)A^I(q,\omega)\Delta N_\omega^{e,i}, \tag{6}$$

$$\Delta N_\omega^{e,i}=N(\omega/T_e)-N(\omega/T_i). \tag{7}$$

The mode occupations $N(\omega/T_\tau)$ are Bose factors.

D. Simplified form of the FGR result

A simplified form of the FGR, presented in detail by Hazak *et al.* [7] is summarized below. If the temperatures and densities are such that $\omega_{\tau,p}/T_i \ll$ unity, the mode-occupation numbers may be expanded as

$$N(\omega/T_e)-N(\omega/T_i)=(T_e-T_i)/\omega. \tag{8}$$

The ion spectral function A^i is limited to the low-energy regime of the ion-plasma frequency, while the electron spectral function A^e lies at the much higher energies of the electron-plasma frequency ω_p^e. Hence, A^e can also be approximated by a frequency-moment expansion about $\omega=0$. Only the first moment is retained. Since $A^e(q,\omega)$ is antisymmetric in ω, we have

$$A^e(q,\omega)=\omega[\partial A^e(q,\omega)/\partial\omega]_{\omega=0}. \tag{9}$$

Hence, the sum over mode frequencies (ω) can be evaluated using the *f*-sum rule for the ionic subsystem.

The result is a simplified FGR formula,

$$R_{f\text{-}sum}=\omega_{ip}\Sigma_{\mathbf{q}}V_q|M_q|^2\left[\mathrm{Im}\frac{\partial\chi^{0e}}{\partial\omega}\right]\Big/\,|\varepsilon(q,0)|^2, \tag{10}$$

where $\varepsilon(q,0)$ is the static electron screening function given by

$$\varepsilon(q,0)=1-V_q(1-\mathcal{G}_q^{ee})\mathrm{Re}\,\chi^0_{ee}(q,0). \tag{11}$$

The $\mathrm{Re}\,\chi^0_{ee}(q,0)$ in $\varepsilon(q,0)$ can be adequately approximated by the $q=0$ value, but the detailed q dispersion of $\mathrm{Im}\chi_{ee}$ at finite T (cf., Ref. [8]) is needed for accuracy. The simplified form mimics the LS formula in containing no details of the ion spectrum, but differs from it in having no $1/T_i^{3/2}$ term, and in invoking the detailed q dispersion of χ_{ee} at low energies. For most three-dimensional plasmas, χ_{ee} near $\omega=0$ is single-particle-like as the collective modes begin at ω_{ep}.

E. The coupled-mode formula

The electron-ion interaction is not particularly weak, especially if T_i is small and $Z>1$. The unscreened e-i coupling constant Γ_{ie} is of the order of $(\Gamma_{ee}\Gamma_{ii})^{1/2}$ and tends to be large in strongly coupled systems. This should not be confused with the usage where the e-i interaction in, say, Al, described by a pseudopotential, is (legitimately) taken to be "weak," when considering the interaction with a ısingle ion. Then the ion temperature does not play a role and U_{ei} is essentially an external potential (e.g., as in a band structure calculation).

The coupled-mode picture is simply the two-component picture of a plasma or a liquid metal where the ion-density fluctuations have become ion-acoustic modes. The electron excitation modes and the ion modes are both determined by the zeros of a single denominator. We have

$$\chi_{\tau\tau}=\chi^0_{\tau\tau}[1-V_{\tau'\tau'}(k)(1-\mathcal{G}_{\tau'\tau'}]\chi^0_{\tau'\tau'}]/\mathcal{D}, \tag{12}$$

$$\mathcal{D}=D_{ee}D_{ii}-D_{ei}, \tag{13}$$

$$D_{\tau\tau}=[1-V_{\tau\tau}1-\mathcal{G}_{\tau\tau})\chi^0_{\tau\tau}], \tag{14}$$

$$D_{ei}=|V_{ei}|^2\chi^0_{ii}\chi^0_{ee}(1-\mathcal{G}_{ie})(1-\mathcal{G}_{ei}). \tag{15}$$

The energy relaxation occurs from hot coupled modes (which are like electron modes if T_e is the higher temperature) to cooler coupled modes (which are like ion modes). The ER rate within the CM picture is given by

$$R_{cm}=\Sigma_{\mathbf{q},\omega}|U_{ie}(q)|^2\omega\frac{\Delta N^{e,i}_{\omega}A^e(q,\omega)A^I(q,\omega)}{|1-|U_{ie}(q)|^2\chi_{ee}\chi_{ii}|^2}. \tag{16}$$

This reduces to the FGR form if the $|U_{ie}(q)|^2$ term which appears in the denominator is negligible compared to unity.

A proof of this formula was given in Ref. [1] using the Keldysh contour technique. The Keldysh approach is excessively abstract, and does not reveal an underlying physical picture. We have succeeded in constructing a simpler, more intuitive derivation which is given in the Appendix.

III. NUMERICAL RESULTS AND DISCUSSION

In Fig. 1 we show the ER coupling constants calculated using the theoretical models discussed in the preceding section. The results are for aluminum at unit compression. Two values of T_i are considered, i.e., $T_i=0.081$ and 10 eV, while the electron temperature is varied. The Landau-Spitzer estimates (dashed curves in the figure) of g_{LS} are virtually independent of T_i, since this appears in LS as $(T_i/M_i+T_e/m_e)^{3/2}$, and is unimportant here. The f-sum evaluation of the FGR yields $g_{f\text{-}sum}$, shown as crosses in the figure. It is completely independent of T_i, in contrast to the Landau-Spitzer form. This is because the f-sum approach is valid only if the ion spectrum lies far below the electron spectrum, while no such restriction applies to the LS formula. The full FGR calculation shows a small dependence on T_i, as shown in the figure (dotted line, $T_i=0.081$ eV; dotted-dashed line, $T_i=10$ eV). In contrast, the coupled-mode result g_{cm} is very sensitive to the T_i. The CM curve at $T_i=0.081$ eV lies much below the curve at $T_i=10$ eV. The change of gradient in the $T_i=0.081$ eV, the CM curve near 10 eV signals the end of the constant $Z=3$ ionization. The sensitivity of the CM description to the ion temperature is clear, since at $T_i=0.081$ eV and $T_e=40$ eV, the electron-ion coupling constant Γ_{ei} is of the order of 33 for Al. The g_{cm} and g_{FGR} are calculated with the *initial* ion distribution held fixed, while T_e is increased. While g_{FGR} is almost independent of the initial ion distribution, g_{cm} depends on the initial conditions set by the experiment.

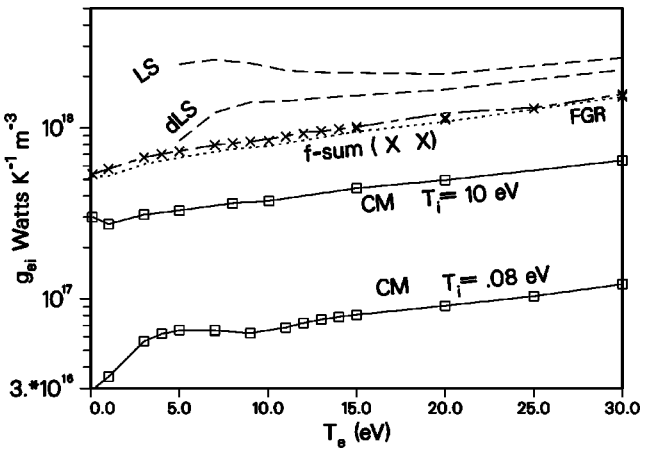

FIG. 1. Electron-ion coupling constant for Al calculated from the theories discussed here are shown for $T_i=0.08$ eV, i.e, melting temperature of Al, and $T_i=10$ eV. The labels LS, DLS, f-sum, FGR, and CM refer to the Landau-Spitzer [Eq. (2)], degeneracy-corrected LS [Eq. (3)], Fermi golden rule [Eq. (6)], f-sum [Eq. (10)], and the coupled-mode form [Eq. (16)], respectively. The LS, DLS curves (dashes) are very weakly dependent on T_i. The f sum (crosses) is independent of T_i. The FGR at $T_i=10$ eV follows the crosses, while the $T_i=0.08$ eV departs slightly from it. The solid line and squares show the CM results.

Table I compares the ER-coupling constants for C, at $T_i=0.5$ eV, i.e., just above its melting point, and for Al and Si with $T_i=0.15$ eV, i.e., just above the melting point of Si. The C and Si pseudopotentials correctly reproduce the short-range structure of the molten elements [9]. The remarkable feature seen here is that the differences in the two elements Al and Si *do not* translate into a big difference in the coupling constants.

Energy relaxation rates for typical elements at different compressions, and for any reasonable range of values of T_e and T_i may be calculated by remotely accessing our computer codes. Interested researchers may do their own calculations via web access [10].

IV. CONCLUSION

We have presented results for the energy-relaxation coupling constants of Al, C, and Si plasmas. The results show that the energy relaxation rate (i.e, from the coupled-mode approach) is probably an order of magnitude smaller (at melting-point temperatures) than those obtained from traditional calculations. The differences between the coupled-

TABLE I. Energy-relaxation coupling constant g_{ER} for Al and Si at 2.7 and 2.59 g/cc, respectively, with $T_i=0.15$ eV (effectively, the meltimg point of Si). For carbon, density is 2.0 g/cc, and $T_i=0.5$ eV, effectively its melting point. Results from the Landau-Spitzer (LS) and coupled-mode (CM) theories are given in $\mathrm{W\,K^{-1}\,m^3}$. The $S(k,\omega)$ is constructed from the hypernetted-chain equation; cf. Ref. [1].

	LS (10^{19} $\mathrm{W\,K^{-1}\,m^{-3}}$)			CM (10^{19} $\mathrm{W\,K^{-1}\,m^{-3}}$)		
T_e	C	Al	Si	C	Al	Si
3	1.4500	0.1998	0.2794	0.3267	0.02024	0.01996
5	1.8053	0.2360	0.3423	0.3326	0.02928	0.03010
10	2.2059	0.2210	0.3968	0.3325	0.03774	0.04885
15	2.2096	0.2079	0.3789	0.3156	0.04419	0.05672
20	1.7490	0.2071	0.2943	0.2960	0.04489	0.05798

mode form and the Fermi golden rule results disappear at sufficiently high ion temperatures.

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APPENDIX: A SIMPLER DERIVATION OF THE COUPLED-MODE FORMULA

The electrons (i.e, effectively the continuum electrons) at temperature T_e, transfer energy to coupled modes (basically the screening electrons closely associated with the ion modes). Their mode-occupation number is denoted by $N_{cm}(\omega)$, depends on both T_e and T_i, and is as yet unspecified. We apply the Fermi golden rule to this process and write the energy transfer rate as

$$R_{e\to cm}=\Sigma_{\mathbf{q},\omega}|U_{ie}(q)|^2A_e(\mathbf{q},\omega)A_{cm}(\omega)\omega\Delta N_{T_e,cm},$$

$$\Delta N_{T_e,cm}=[N(\omega/T_e)-N_{cm}(\omega)].$$

The spectral function $A_{cm}(\omega)=-2\,\mathrm{Im}\,\chi_{ii}$. This involves the coupled-mode response function for ions, viz., Eq. (12). Similarly, the transfer of energy from the coupled modes to the ion subsystem (i.e, basically the nucleii and core electrons) is

$$R_{cm\to i}=\Sigma_{\mathbf{q},\omega}|U_{ie}(q)|^2A_i(\mathbf{q},\omega)A_{cm}\omega\Delta N_{cm,T_i},$$

$$\Delta N_{cm,T_i}=[N_{cm}(\omega)-N(\omega_{\mathbf{q}}/T_i)].$$

When a steady state is reached, the rates of transfer $R_{e\to cm}$ and $R_{cm\to i}$ become equal:

$$R_{cm}=R_{e\to cm}=R_{cm\to i}. \tag{A1}$$

This arises when the coupled-mode population becomes

$$N_{cm}(\omega)=\frac{\overline{N}(\omega/T_i)A^i(q,\omega)+\overline{N}(\omega/T_e)A^e(q,\omega)}{A^i(q,\omega)+A^e(q,\omega)}. \tag{A2}$$

This is Eq. (46) of Ref. [1] obtained there using a nonequilibrium Green's-function technique based on the Keldysh contour integration. The present physical picture is not needed in the Green's-function approach. It follows that under steady-state conditions, the energy relaxation rate is given by R_{cm}, which is evaluated using the above expression for N_{cm}. This easily reduces to Eq. (50) of Ref. [1]. Thus we see that coupled modes, being hotter than bare ions, and colder than bare electrons, create an energy-relaxation bottleneck.

[1] M.W.C. Dharma-wardana and F. Perrot, Phys. Rev. E **58**, 3705 (1998); **63**, 069901 (2001).
[2] L. Spitzer, *Physics of Fully Ionized Gases* (Interscience, New York, 1962).
[3] D. Riley, N.C. Woolsey, D. McSherry, I. Weaver, A. Djaoui, and E. Nardi, Phys. Rev. Lett. **84**, 1704 (2000); P. Cellier, A. Ng, G. Xu, and A. Forsman, *ibid.* **68**, 2305 (1992); Phys. Rev. E **52**, 4299 (1995); P.B. Corkum, F. Brunel, N.K. Sherman, and T. Srinivasan-Rao, Phys. Rev. Lett. **61**, 2886 (1988).
[4] F. Perrot and M.W.C. Dharma-wardana, Phys. Rev. B **62**, 16 536 (2000).
[5] Y.T. Lee and R.M. More, Phys. Fluids **27**, 1273 (1984).
[6] H. Brysk, Plasma Phys. **16**, 927 (1974).
[7] G. Hazak, Z. Zinamon, Y. Rosenfeld, and M.W.C. Dharma-wardana (unpublished).
[8] F.C. Khanna and H.R. Glyde, Can. J. Phys. **54**, 648 (1976).
[9] M.W.C. Dharma-wardana and F. Perrot, Phys. Rev. Lett. **65**, 76 (1990).
[10] See http://nrcphy1.phy.nrc.ca/ims/qp/chandre/ and follow the link from NEWSTUFF, or go directly to: http://nrcphy1.phy.nrc.ca/ims/qp/chandre/Lst.