

Thermalization of an electron-phonon system in a nonequilibrium state characterized by fractal distribution of phonon excitations

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(Received 5 December 1996)

The thermalization of an electron-phonon system in a nonequilibrium state characterized by a fractal distribution of phonon excitations is calculated in a novel way, which takes into account the unusual, inverse power-law type distribution of phonon energies. The calculations are done from the first principles, based on a nonequilibrium statistical operator combined with the recently proposed generalized, nonextensive thermostatics. As a result the usual linear (Newtonian) energy transfer rate $\dot{Q} \propto -(T_p - T_e)$ is replaced by a nonlinear rate $\dot{Q} \propto -(T_p^q - T_e^q)$, where $q \geq 1$ is related to the fractality of the excitation energy distribution. Consequences of this result for a thermal relaxation of the nonequilibrium phonon system are discussed. [S1063-651X(97)13506-1]

PACS number(s): 05.20.-y, 63.20.Kr, 5.70.Ln, 61.85.+p

The thermalization of a nonequilibrium electron-phonon system is of interest in diverse fields of physics, such as the materials modifications by energetic ion beams or by intense laser irradiation. The modeling of the thermalization is usually treated with coupled nonlinear heat transport equations for electron (e) and phonon (p) subsystems, where the energy transfer rate \dot{Q} from the phonons to electrons is described by a Newtonian cooling term $\dot{Q} \propto -(T_p - T_e)$ proportional to the temperature difference between the subsystems [1-4]. The Newtonian cooling term is an outcome of all established theories of the thermal relaxation of nonequilibrium electron-phonon systems [5,6] and its applicability in describing the nonequilibrium states has not been seriously questioned, although the restrictions of the theory are well known [6].

Fractal or inverse power-law type distributions of phonon excitations are of interest in modeling the thermalization of collision cascades in ion bombarded solids, where a microscopic distribution of energies of lattice ions deviate substantially from the thermal or Boltzmann-Gibbs (BG) distribution [7,8]. The deviations are apparent even at a stage, where all atoms are moving [7] and the atomistic motion is expected to be correlated, giving rise to localized phonon-type excitations. The relevance of phonon-type excitations in the thermalization of cascade is indirectly supported by the notion that the thermally activated radiation enhanced processes in metals correlate strongly with the electron-phonon coupling strength (for a more detailed discussion, modeling and comparisons with experiments, see Refs. [1,2]). Instead of being described by the BG distribution the energy distribution of atoms fits better to an inverse power law of a form $p(\epsilon) \propto \epsilon^{-\alpha}$, where $1 \leq \alpha \leq 2$ [7,8]. This is to be expected, because the thermalization can be treated as an energy sharing branching process, which has much in common with a fragmentationlike behavior, where a self-similar (inverse power-law) distribution of fragments is produced [7-9].

Until recently there has been little guidance on how to generalize the theory of thermalization for an electron-phonon systems obeying non-Gibbsian statistics. The task of generalization can now be accomplished within the framework of recently proposed generalized nonextensive (Tsallis) thermostatics [10,11], where the Gibbs ensemble is replaced by a more general Tsallis' ensemble $p(\epsilon) \propto [1 - \beta(1-q)\epsilon]^{-1/(q-1)}$, where $q \geq 1$ and β is a generalized inverse temperature (actually a Lagrange multiplier appearing in the maximization of the entropy). The parameter defining the deviation from the ordinary statistics is the Tsallis index q , which is related to the nonextensivity (or fractality) of the system [11]. In the limit $q \rightarrow 1$ the ordinary Gibbs ensemble is obtained. Because the Tsallis' ensemble bridges the BG distribution and inverse power-law distributions, it is adapted in describing the statistics of the thermalizing cascade, where typical values of Tsallis index fall between the limits $1.5 \leq q \leq 1$. Of course, even the Tsallis' statistics is too idealized to give a completely realistic description of the thermalization of the collision cascade, but it is, nevertheless, an improvement over the all too restrictive BG statistics. In this study the established results of the thermalization of electron-phonon system are rederived within the framework of Tsallis' statistics. The exposition is brief, and many details, not of immense interest for the present study, are omitted by giving a reference to original works, where a more complete discussion can be found.

The microscopic model of the electron-phonon system is characterized by a Hamiltonian $\hat{H} = \hat{H}_e + \hat{H}_p + \hat{U}_{ep}$, where electron and phonon systems are described by terms $\hat{H}_e = \sum_{\mathbf{k}} \epsilon_{\mathbf{k}} a_{\mathbf{k}}^\dagger a_{\mathbf{k}}$ and $\hat{H}_p = \sum_{\mathbf{Q}} \hbar \omega_{\mathbf{Q}} b_{\mathbf{Q}}^\dagger b_{\mathbf{Q}}$, respectively, and coupled by the Fröhlich-type interaction $\hat{U}_{ep} = \sum_{\mathbf{Q}, \mathbf{k}} M_{\mathbf{Q}} (b_{\mathbf{Q}} + b_{-\mathbf{Q}}^\dagger) a_{\mathbf{k}}^\dagger a_{\mathbf{k}-\mathbf{Q}}$ [6]. The transition matrix $M_{\mathbf{Q}}$ gives the probability for a scattering process, where the momentum and energy of the electron are changed from \mathbf{k} to $\mathbf{k}-\mathbf{Q}$ and $\epsilon_{\mathbf{k}}$ to $\epsilon_{\mathbf{k}-\mathbf{Q}}$, respectively, accompanied by the creation or destruction of the phonon (or density fluctuation) with energy $\hbar \omega_{\mathbf{Q}}$. The energy current operator between the subsystems in the nonequilibrium state is obtained from the quantum mechanical equation of motion

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$$\begin{aligned}\hat{J} &= (i\hbar)^{-1} [\hat{H}_p + \hat{U}_{ep}, \hat{H}] \\ &= (i\hbar)^{-1} \sum_{\mathbf{Q}, \mathbf{k}} M_{\mathbf{Q}} (\epsilon_{\mathbf{k}} - \epsilon_{\mathbf{k}-\mathbf{Q}}) (b_{\mathbf{Q}} + b_{-\mathbf{Q}}^\dagger) a_{\mathbf{k}}^\dagger a_{\mathbf{k}-\mathbf{Q}}.\end{aligned}\quad (1)$$

The energy transfer rate \dot{Q} is then the expectation value of operator \hat{J} over the suitable statistical ensemble described by density operator $\hat{\rho}$, which can be constructed according to Zubarev's theory [12,13]. This method based on the nonequilibrium statistical operator is preferred instead of the approach based on the kinetic equations, because it is more transparent with respect to the central approximations needed to reach the final results. The generalization of the Zubarev's nonequilibrium statistical operator compatible with Tsallis' statistics is now given by

$$\hat{\rho}_q(\hat{A} + \hat{B}) = Z_q^{-1} [\hat{1} - (1-q)(\hat{A} + \hat{B})]^{-1/(q-1)}. \quad (2)$$

The part of the operator which defines the equilibrium is $\hat{A} = \beta_e(\hat{H}_e - \mu\hat{N}_e) + \beta_p\hat{H}_p$, where the inverse temperatures of the subsystems are $\beta_{e,p} = 1/kT_{e,p}$. The chemical potential of the fermion system with the number operator $\hat{N}_e = a_{\mathbf{k}}^\dagger a_{\mathbf{k}}$ is taken to be $\mu = E_F$. The part

$$\hat{B} = \lim_{\varepsilon \rightarrow 0} \int_{-\infty}^0 dt e^{\varepsilon t} (\beta_e - \beta_p) \hat{J}(t) \quad (3)$$

of the density operator is due to the nonequilibrium energy current $\hat{J}(t)$ (in the Heisenberg representation). This expression is valid for a weak interaction but for an arbitrarily large temperature difference [12]. The partition function is given now by $Z_q(\hat{A} + \hat{B}) = \text{Tr}[\hat{1} - (1-q)(\hat{A} + \hat{B})]^{-1/(q-1)}$, where q defines the degree of fractality or (nonextensivity) of the system [14]. In the limit $q \rightarrow 1$ density operator takes the usual form $\hat{\rho} = Z^{-1} \exp[-\hat{A} - \hat{B}]$ corresponding to the Gibbs ensemble.

The energy transfer rate \dot{Q} is the expectation value of the operator \hat{J} , and in the Tsallis' statistics this expectation value is given by [14]

$$\dot{Q} \equiv \langle \hat{J} \rangle_q \equiv \text{Tr}[\hat{J} \hat{\rho}_q(\hat{A} + \hat{B})]. \quad (4)$$

A crucial part of the calculation is now a decomposition of the statistical operator $\hat{\rho}_q(\hat{A} + \hat{B})$ in a similar way as done by the Kubo identity when the Gibbs ensemble is used (see, e.g., Refs. [12,13]). The decomposition in the Tsallis' statistics can be accomplished by using generalized Kubo identities due to Rajagopal [15], yielding

$$\dot{Q} = q \text{Tr} \left[\hat{J} \hat{\rho}_q^q(\hat{A}) \int_0^1 d\lambda \hat{\rho}_q^{-q}(\lambda \hat{A}) \hat{B}_\lambda \hat{\rho}_q^q(\lambda \hat{A}) \right], \quad (5)$$

where $\hat{B}_\lambda = [\hat{1} - \lambda(1-q)\hat{A}]^{-1} \hat{B} [\hat{1} - \lambda(1-q)\hat{A}]^{-1}$. At the limit $q \rightarrow 1$, Eq. (5) becomes identical with the result obtained from the usual Kubo identity (compare with calculations in Refs. [12,13]). The operator product depending on λ can be expanded in the form $\hat{\rho}_q^{-q}(\lambda \hat{A}) \hat{B}_\lambda \hat{\rho}_q^q(\lambda \hat{A}) \approx e^{-\lambda \hat{A}} [\hat{1} + O(\hat{A}^2)] \hat{B} [\hat{1} - O(\hat{A}^2)] e^{\lambda \hat{A}}$, where $O(\hat{A}^2)$ denotes

operators, which depend on \hat{A}^2 or higher order terms. Because \hat{J} is of the order $O(\hat{A}^2)$ itself, we can neglect the terms $O(\hat{A}^2)$ in the expansion, when we are interested in dilute excitations only (this assumption is also behind all the kinetic formulations as, e.g., in Ref. [6]). In practice, this assumption greatly simplifies the calculations by retaining the structure of the ordinary Kubo identity in Eq. (5). Performing then the integration over λ one obtains

$$\begin{aligned}\dot{Q} &= -(i\hbar)^{-1} \lim_{\varepsilon \rightarrow 0} \int_{-\infty}^0 dt e^{\varepsilon t} (\beta_e - \beta_p) \sum_{\mathbf{k}, \mathbf{Q}} \sum_{\mathbf{k}', \mathbf{Q}'} M_{\mathbf{Q}} M_{\mathbf{Q}'} \\ &\times \frac{(\epsilon_{\mathbf{k}} - \epsilon_{\mathbf{k}-\mathbf{Q}})(\epsilon_{\mathbf{k}'} - \epsilon_{\mathbf{k}'-\mathbf{Q}'})}{\beta_e(\epsilon_{\mathbf{k}} - \epsilon_{\mathbf{k}-\mathbf{Q}}) + \beta_p \hbar \omega_{\mathbf{Q}}} \text{Tr} \{ \hat{\rho}_q^q(\hat{A}) \\ &\times [(b_{\mathbf{Q}} + b_{-\mathbf{Q}}^\dagger) a_{\mathbf{k}}^\dagger a_{\mathbf{k}-\mathbf{Q}}, e^{iHt/\hbar} (b_{\mathbf{Q}'} + b_{-\mathbf{Q}'}^\dagger) \\ &\times a_{\mathbf{k}'}^\dagger a_{\mathbf{k}'-\mathbf{Q}'} e^{-iHt/\hbar}] \},\end{aligned}\quad (6)$$

where $\text{Tr}\{\dots\}$ is actually a temperature ordered Green's function (for technical details in similar calculations see Refs. [12,13]). It is now possible to proceed in the usual way [12,13], by pairing the operators according to Wick's theorem, neglecting the interaction term \hat{U}_{ep} in the time-evolution operators, performing the integration and then taking the limit $\varepsilon \rightarrow 0$. In this procedure, the inverse temperature difference $\beta_e - \beta_p$ is canceled by the denominator $[\beta_e(\epsilon_{\mathbf{k}} - \epsilon_{\mathbf{k}-\mathbf{Q}}) + \beta_p \hbar \omega_{\mathbf{Q}}]$, when the energy conservation $\epsilon_{\mathbf{k}} - \epsilon_{\mathbf{k}-\mathbf{Q}} = \hbar \omega_{\mathbf{Q}}$ is taken into account. The final result, after rearranging the terms in the remaining double sum, can be written in a form

$$\begin{aligned}\dot{Q} &= -\frac{4\pi}{\hbar} \sum_{\mathbf{k}, \mathbf{Q}} \hbar \omega_{\mathbf{Q}} |M_{\mathbf{Q}}|^2 f_{\mathbf{k}} (1 - f_{\mathbf{k}-\mathbf{Q}}) [n_{\mathbf{Q}} \delta(\epsilon_{\mathbf{k}} - \epsilon_{\mathbf{k}-\mathbf{Q}} \\ &+ \hbar \omega_{\mathbf{Q}}) - (1 + n_{\mathbf{Q}}) \delta(\epsilon_{\mathbf{k}} - \epsilon_{\mathbf{k}-\mathbf{Q}} - \hbar \omega_{\mathbf{Q}})],\end{aligned}\quad (7)$$

where $f_{\mathbf{k}}$ and $n_{\mathbf{Q}}$ are occupation probabilities of quantum many-body systems for fermions and bosons, respectively, to be calculated in the Tsallis' statistics. However, at present the exact expressions in closed form for these distributions are not known, and results only in the form of Hillhorst integral transformations are available [16]. Fortunately, at this stage of the calculation, it is enough to be able to prove that also in the Tsallis' statistics, when deviations from BG statistics are moderate and $q \approx 1$, the well-known relations $f(\epsilon \pm \hbar \omega) - f(\epsilon) \approx \hbar \omega \delta(\epsilon - E_F)$ and $f(\epsilon \pm \hbar \omega) [1 - f(\epsilon)] \approx n(\omega) [f(\epsilon) - f(\epsilon \pm \hbar \omega)]$ hold. This can be done, for example, by using mean fieldlike expansions for $f_{\mathbf{k}}$ and $n_{\mathbf{Q}}$ derived by Büyükkılıç, Demirhan, and Gülec [17] (however, for criticism see [16,18]). With the above relations the leading contribution of the energy transfer rate can be obtained by following the method of Allen [6], first passing in the continuum limit and then introducing the electron-phonon spectral function $\alpha^2 F(\omega) = [\hbar N(E_F)/2]^{-1} \sum_{\mathbf{k}, \mathbf{k}'} |M_{\mathbf{Q}}|^2 \delta(\omega_{\mathbf{Q}} - \omega) \times \delta(\epsilon_{\mathbf{k}} - \epsilon) \delta(\epsilon_{\mathbf{k}'} - \epsilon)$ (also known as Eliashberg's function), where $\mathbf{k} - \mathbf{k}' = \mathbf{Q}$ and $N(E_F)$ is the density of states of the electrons at the Fermi level. Then Eq. (7) can be cast in a form

$$\dot{Q} = -2\pi N(E_F) \int d\omega \alpha^2 F(\omega) (\hbar\omega)^2 [n(\omega, T_L) - n(\omega, T_e)]. \quad (8)$$

In a further evaluation of Eq. (8) it is now necessary to adopt an approximate expression for the phonon distribution $n(\omega, T)$. A recent work of Tsallis *et al.* [11] on a generalized Bose-Einstein distribution in a nonextensive statistics provides for a phonon distribution the following convenient approximation ,

$$n(x) = n_{BE}(x)(1 - e^{-x})^{q-1} \left\{ 1 + (1-q)x \left[\frac{1 + e^{-x}}{1 - e^{-x}} - \frac{x}{2} \frac{1 + 3e^{-x}}{(1 - e^{-x})^2} \right] \right\}, \quad (9)$$

where $n_{BE}(x) = [\exp(x) - 1]^{-1}$, with $x = \hbar\omega/k_B T$, is the usual Bose-Einstein distribution. This distribution has an advantage of being representable in a form of converging series, which allows us to find an approximation to \dot{Q} also in a form of converging series. It becomes now evident, that although Eq. (8) is formally identical to the Allen's Eq. (10), its high temperature expansion is quite different. Performing the expansion in terms $x \ll 1$, changing in dimensionless variables $\tilde{T} = T/\Theta_D$, where Θ_D is the Debye temperature, the leading contribution of the energy transfer rate is obtained

$$\dot{Q} \approx -\pi \hbar^2 N(E_F) \omega_D \lambda_q \langle \omega^2 \rangle (\tilde{T}_p^q - \tilde{T}_e^q) \left(1 - C \frac{1}{12} \frac{1}{\tilde{T}_p \tilde{T}_e} \right), \quad (10)$$

where ω_D is the Debye frequency. The generalized coupling strength is defined as $\lambda_q \langle \omega^2 \rangle = (\bar{\Omega}^{3-q}/\bar{\Omega}^2) \lambda \langle \omega^2 \rangle$, where $\lambda \langle \omega^2 \rangle$ is the standard coupling strength factor defined as in Ref. [6]. The dimensionless (generalized) moments $\bar{\Omega}^r = 2 \int_0^\infty d\Omega [\alpha^2 F(\Omega)/\Omega] \Omega^r$, where $\Omega = \omega/\omega_D$, appearing in the definition of the coupling strength are only numerical correction factors which can be calculated once the phonon spectrum is known. The coefficient $C = [(1-q) + q(3q-1)/2] \bar{\Omega}^4/\bar{\Omega}^2$ in the second term in the high temperature expansion is also defined by these dimensionless moments. At the limit $q \rightarrow 1$ Eq. (10) is identical with the Allen's results [6], but in general case $q > 1$, which means that the energy transfer rate is enhanced in comparison with the standard linear result. The energy transfer rate \dot{Q} is the main result of this study, and it is directly applicable in those analytical studies and molecular dynamics simulations, where energy transfer from phonons to electrons are incorporated as a part of the model calculations [1–3]. However, deviations from the BG statistics must not be too large.

Finally, we estimate the cooling rate of the phonon system initially heated up by the impact of an energetic ion. For simplicity, the electronic subsystem is treated as a heat sink with $T_e \ll T_p$, and the contribution of the ionic heat conduction on the cooling is ignored, which are reasonable approximations for large and sparse cascades [2]. However, in the modeling aiming at accurate predictions, the contribution of the heat conduction on the cooling must be taken into account, but then it is best to deal with the established simula-

tion models [1,3]. Under these assumptions, the cooling rate of the phonon system is simply due to electron-phonon coupling and it is given by $(\partial T_p/\partial t)_{ep} = \dot{Q}/C_q$, where C_q is the heat capacity of the phonon system with Tsallis index q . Because we are now concerned with a model of independent, localized oscillators at a high temperature limit, the existing calculations for a heat capacity of one oscillator [19,20] can be used as a guidance. Towards this end the results due to Guerberoff, Pury, and Raggio [19] for the heat capacity are adopted. The series they give for the heat capacity (on p. 1801 in Ref. [19]) can be approximated by corresponding integrals, yielding for one oscillator at high temperatures $C_q \propto (2-q)^q T^{1-q}$ [21]. This result suggests that the heat capacity can be approximated by $C_q \approx C_{DP} (2-q)^q \tilde{T}_p^{1-q}$, where $C_{DP} = 3Nk$ is the Dulong-Petit heat capacity. The estimate has an advantage in that it reproduces the temperature dependence of the heat capacity of the classical ideal gas in the Tsallis' statistics [22]. By using it, we can write down the temperature relaxation rate of the phonon system as $(\partial \tilde{T}_p/\partial t)_{ep} = -\Gamma \tilde{T}_p^{2q-1}$, where the rate coefficient $\Gamma = \Gamma_0 [\Omega^{3-q}/\Omega^2 (2-q)^q]$ is expressed in terms of the corresponding coefficient $\Gamma_0 = \pi \hbar N(E_F) \lambda \langle \omega^2 \rangle / (C_{DP}/k)$ for the electron-phonon system obeying BG statistics. Values of Γ_0 calculated for ordinary metallic systems are given, e.g., in Ref. [2]. We have thus ended up with a manifestly nonlinear cooling rate instead of the usual linear rate $(\partial \tilde{T}_p/\partial t)_{ep} = -\Gamma_0 \tilde{T}_p$. The ordinary linear result is now understood to be the limiting case for $q \rightarrow 1$, and thus valid only in a completely thermalized stage.

The validity of the derived heat transfer rate \dot{Q} and the temperature relaxation rate $(\partial T_p/\partial t)_{ep}$ are both restricted to small deviations from BG statistics, when $q \approx 1$. This limits the applicability of the results at the early stage of the cascade thermalization, where values as large as $q \approx 1.5$ may be attained. At present the direct verification of the energy transfer or cooling rates for the nonequilibrium phonon system in collision cascades is not possible, because the thermalization is over in picoseconds [1,2]. Therefore, the value of the results must be judged on the basis of their theoretical consistency with the supposed initial conditions. In any case, it is interesting to see, how the adoption of the statistical ensemble, where the most restrictive assumptions are known to culminate, has so crucial a role in determining the energy transfer rate. It is now seen, how the Newtonian cooling term $\dot{Q} \propto -(T_p - T_e)$ is basically a consequence of the restrictions imposed by the adopted ensemble (note that the present derivation demonstrates also how the Newtonian cooling term is obtained without assuming a small temperature difference between the subsystems). We should be reminded that the fractal distribution of phonon excitations studied here also represents an idealization. This idealized system is not meant to replace the models based on the BG statistics but instead to give a complementary point of view, which gives more insight on the role of the anomalous statistics in the thermalization problem. A realistic model should probably describe the relaxation by a sequence of Tsallis' ensembles starting initially from a value $q \approx 1.5$ and ending up with a value $q = 1$ (see, e.g., results in Ref. [7]), possibly by means of

time dependent parameter q determined by the dynamics of the collision cascade. A similar situation arises in the evolution of cascade geometry, where initially fractal cascade evolves towards space filling structure with time dependent fractal index [8,23].

In summary, in this paper the generalized energy transfer rate from phonons to electrons is derived for systems, where phonon excitations has an anomalous, inverse power-law type energy spectrum. This model has potential applications in the description of the thermalizing collision cascades or energy sharing branching processes. The generalized rate derived here has a theoretical basis reaching beyond the as-

sumed validity of the Boltzmann-Gibbs distribution (i.e., completely thermalized state). Due to the fractality of the phonon distribution, the rates of the energy transfer and the temperature relaxation of the phonon system are enhanced in comparison with the standard results. The results demonstrate the profound effects of the assumptions contained in the choice of the statistical ensemble.

The author wishes to thank Professor C. Tsallis, Dr. P. Pury, and Dr. G. Raggio for comments and helpful correspondence. J. Peisa is acknowledged for his kind help in cross-checking part of the calculations.

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