Instability of Metals with Respect to Strong Electron-Phonon Interaction

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We show that thermal equilibrium between conduction electrons and phonons becomes kinetically unstable when the renormalized electron-phonon coupling exceeds a certain threshold. We prove that negative electronic specific heat, $C_{\rm el} < 0$, is sufficient to trigger the instability. Specifically, the instability sets in as soon as the quasiparticle weight becomes negative over a range of energies, even before $C_{\rm el}$ turns negative. This is an inherently nonequilibrium phenomenon, occurring prior to the formation of any equilibrium phase. Depending on the system, it can proceed along different pathways, ultimately resulting in a structural transition to an insulating or metallic state.

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Although the idea that the strength of the *physical* (renormalized) electron-phonon interaction in metals is somehow limited by a lattice instability has been around for more than half a century, a clear understanding of whether a fundamental upper bound of this kind truly exists is sill lacking [1–3]. On one hand, it is well known that the metallic phase of the electron-phonon models, such as Holstein, Frölich, Su-Schrieffer-Heeger, etc., becomes unstable when the *bare* (unrenormalized) dimensionless electron-phonon coupling $\lambda_0 > 0.5$ [4–7]. The instability is variably described in the literature as lattice reconstruction or charge-density wave or as polaronic or bipolaronic transition [7–19]. On the other hand, the physical electron-phonon coupling λ, which determines the superconducting T_c and other physical properties, diverges at the instability [4–6]. Since $T_c \propto \sqrt{\lambda}$ for large λ [20–25], T_c too can take arbitrarily large values.

The situation is further complicated by the fact that the electron-phonon models cannot be consistently derived from the underlying Hamiltonian of electrons and ions interacting via Coulomb forces and, therefore, do not provide a fully accurate description [26–31]. When supplemented with the actual phonon spectrum, they do capture the effects due to phonon mediated electron-electron interaction [26–29,32]. Yet it is impossible to obtain the correct phonon spectrum from within these models. The main reason is double counting (also known as overscreening) electronic polarization in the renormalized phonon spectrum [3,26,27,30,31]. This contribution is already taken into account once when obtaining, e.g., the Holstein or Frölich models from the electron-ion Hamiltonian, and the subsequent calculation of the phonon spectrum within these models includes it once more. As a result, physical phonon frequencies $\omega_{\boldsymbol{k}} \approx \omega_{\boldsymbol{k}0}\sqrt{1-2\lambda_0}$, where $\omega_{\boldsymbol{k}0}$ are the bare frequencies, soften at $\lambda_0 = 0.5$. A spurious lattice instability, absent in the original electron-ion Hamiltonian, develops, while the renormalized electron-phonon interaction constant $\lambda = \lambda_0/(1-2\lambda_0)$ diverges as $\lambda_0 \to 0.5$. In many cases, the polaronic and charge-density wave (P/CDW) instability found in the electron-phonon models is simply this artificial phonon softening [19].

Most importantly, regardless of whether or not we accept the P/CDW instability at $\lambda_0 \approx 0.5$ as real, we see that it does not lead to well-defined upper bounds on λ and T_c.

In this Letter, we identify a *kinetic instability in metals* that sets a strict upper bound on the *physical* electron-phonon interaction strength. This instability is *fundamentally distinct* from the P/CDW instability also because it is a *nonequilibrium phenomenon*, does *not* involve phonon softening, and is *not* based on effective electron-phonon models. Previous work demonstrated that the electronic specific heat, $C_{\rm el}$, becomes negative when λ exceeds a certain threshold [33–35]. However, since electrons are strongly coupled to phonons, it is not immediately obvious that this necessarily leads to an instability. Additionally, because the phonon contribution to the total specific heat is much larger than $|C_{\rm el}|$ [36], the overall specific heat remains positive [33,34].

Here, we *prove rigorously* that thermal equilibrium between electrons and phonons becomes *intrinsically unstable whenever* $C_{\rm el} < 0$. More importantly, we find that metals *undergo a kinetic instability even before* $C_{\rm el}$ turns negative—this occurs as soon as the quasiparticle weight, $Z_0(E)$ (also known as the wave function renormalization constant), becomes negative over a finite energy

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range. This is an *absolute instability*, meaning the system is unstable to *infinitesimal perturbations* around equilibrium, and it is a fundamental feature of any fermionic system with a retarded interaction mediated by virtual bosons. Instabilities of this kind signal a *first-order phase transition*—for example, the negative compressibility of a van der Waals gas indicates a liquid-gas transition. Here, we anticipate that the system undergoes a *structural transition* into an insulator or another metal [33,35].

The kinetic equation for the electron distribution function $f(E,t)$ in the normal state in the presence of electron-phonon interactions is [38,39]

$$(1-\Sigma')\dot{f}+f'\dot{\Sigma}=I_{\rm ep}, \tag{1}$$

where the dot and prime denote derivatives with respect to time and energy. $I_{\rm ep}$ is the electron-phonon collision integral,

$$I_{\rm ep}=2\pi\int_0^\infty d\omega\alpha^2F(\omega)[N_0(T_0)(f_++f_--2f) - f(f_+-f_-)+f_+-f], \tag{2}$$

$f_\pm=f(E\pm\omega)$, and

$$\alpha^2F(\omega)=\sum_k\frac{\lambda_k\omega_k}{2}\delta(\omega-\omega_k) \tag{3}$$

is the Eliashberg function. All phonon frequencies satisfy $\omega_k>0$, so our treatment does not involve phonon softening. Since the electron-phonon interaction enters through a Fermi surface average in $\alpha^2F(\omega)$, its detailed momentum dependence plays no role in the electron dynamics at leading order in Ω/E_F [37], where E_F is the Fermi energy and Ω the maximum phonon frequency.

The electron self-energy is

$$\Sigma[f]=\fint dx\int_0^\infty d\omega\alpha^2F(\omega)\frac{f(x+\omega)-f(x-\omega)}{E-x}, \tag{4}$$

$N_0(T_0)=[e^{(\omega/T_0)}-1]^{-1}$ is the Bose distribution at temperature T_0, ω_k are phonon frequencies, and λ_k are the coupling constants. This kinetic equation has been derived within the same approximation as the Migdal–Eliashberg (ME) theory [40]. Its r.h.s. explicitly takes into account the electron self-energy due to the electron-phonon interaction, while the direct electron-electron interactions are included through the effective parameters of the underlying Fermi liquid. We also took spatially uniform initial conditions, so that the electron and phonon distribution functions are independent of the spatial coordinates, and assumed that phonons are in thermal equilibrium. The latter is an excellent approximation, because $(C_{\rm ph}/|C_{\rm el}|)\sim(E_F/\Omega)$ [36]. Since the small parameter in the ME theory is $(\Omega/E_F)\to 0$, $C_{\rm ph}\to\infty$ in the limit where this theory is exact. This means that phonons act as a thermal bath with an infinite heat capacity for the fermions [40].

The fixed point of Eq. (1) is the equilibrium Fermi distribution $f_0(E,T_0)=[e^{(E/T_0)}+1]^{-1}$. In equilibrium, $1-\Sigma'[f_0]\equiv Z_0^{-1}(E)$ has the meaning of the inverse quasiparticle weight function. Explicitly [37],

$$Z_0^{-1}=1-(2\pi T_0)^{-1}\int d\omega\,\alpha^2F(\omega)\,{\rm Im}(\psi_+^{(1)}-\psi_-^{(1)}), \tag{5}$$

where $\psi_\pm^{(1)}=\psi^{(1)}\{\frac{1}{2}+[(iE\pm i\omega)/2\pi T_0]\}$ and $\psi^{(1)}(x)$ is the trigamma function. Let us also introduce the dimensionful (g) and dimensionless (λ) electron-phonon coupling constants,

$$g^2=2\int d\omega\,\omega\alpha^2F(\omega)=\sum_k\lambda_k\omega_k^2, \tag{6}$$

$$\lambda=2\int d\omega\frac{\alpha^2F(\omega)}{\omega}=\sum_k\lambda_k. \tag{7}$$

We first analyze the strong coupling limit $\lambda\to\infty$, where the instability is most pronounced. Another attractive feature of this limit is its universality: since $\lambda\to\infty$ is equivalent to taking all phonon frequencies to zero, the results are independent of the phonon dispersion [23]. Finite differences turn into derivatives, e.g., $f_+-f_-\to 2f'\omega$, and the r.h.s. of Eq. (1) becomes $\pi g^2(T_0f''-2ff'+f')$. It is convenient to work in energy units $g=1$ and to rescale the time $\pi t\to t$. In these units, $T_c=0.1827$ in the limit $\lambda\to\infty$ [20]. For small deviations from equilibrium and $E\gg T$, the $f'\dot{\Sigma}$ term on the l.h.s. of Eq. (1) is negligible and $1-\Sigma'\approx Z_0^{-1}$ to the leading order in the deviation. Equation (1) therefore takes the form

$$Z_0^{-1}\dot{f}=T_0f''-2ff'+f'. \tag{8}$$

In terms of new variables $u=2f-1$ and $y=E-\Sigma[f_0]$, this is the well-known Burgers' equation [41,42] with energy (position) dependent viscosity $T_0Z_0^{-1}(E)$,

$$\dot{u}=\frac{d}{dy}\left(T_0Z_0^{-1}\frac{du}{dy}\right)-u\frac{du}{dy}. \tag{9}$$

The $2ff'$ term is similarly negligible for $E\gg T$ and therefore $u(\partial u/\partial y)\approx-(\partial u/\partial y)$. With this replacement, Eq. (9) becomes the 1D convection-diffusion equation with position-dependent diffusion coefficient $T_0Z_0^{-1}(E)$. The expression (5) for $Z_0^{-1}(E)$ simplifies in the strong coupling limit as well [37]. Plotting it for various T_0, we observe that for $T_0\le 1.63T_c$ there is a region where it dips below the horizontal axis. In particular, $Z_0(E)<0$ for $0.40<E<1.16$ at $T_0=T_c$.

The existence of a region of negative viscosity in Burger's equation (9) or, equivalently, negative diffusion coefficient in the diffusion-convection equation indicates

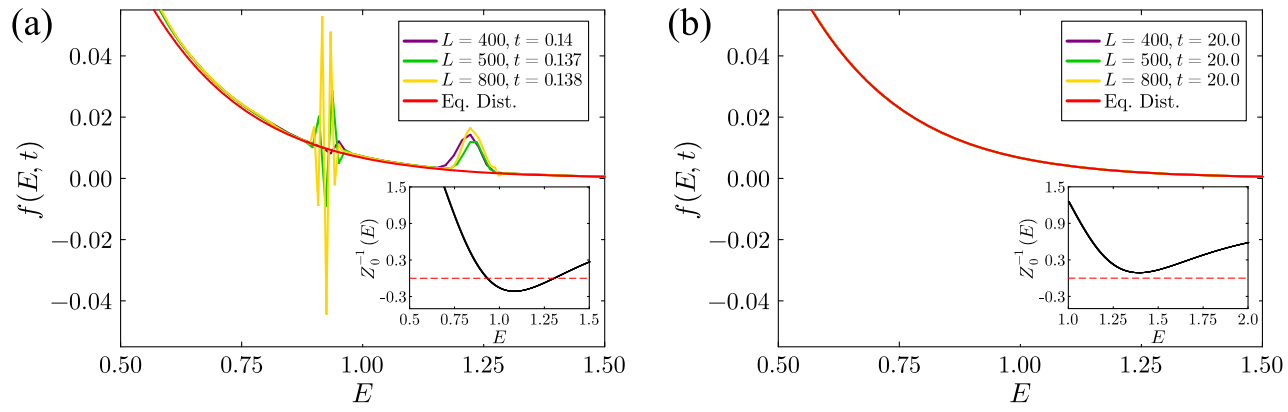

FIG. 1. Instability in the full kinetic equation. We simulate Eq. (1) for Einstein phonons with (a) $\lambda = 2.169 > \lambda_c = 1.27$ and (b) $\lambda = 1.0 < \lambda_c$. The initial electron temperature $T_{\rm in} = 0.202$ is slightly above the phonon temperature $T_0 = 0.20$. Insets show the corresponding inverse quasiparticle weight $Z_0^{-1}(E)$. The solid red line in both panels is the equilibrium Fermi distribution $f_0(T_0)$. The unstable growth of the electron distribution function $f(E,t)$ in (a) is most prominent near the energies $E = 0.94$ and 1.30 satisfying $Z_0^{-1}(E) = 0$. Because of the highly oscillating behavior, the simulation can run only for a short time in (a). There is no instability in (b), since $Z_0^{-1}(E) > 0$, and the solution converges to $f_0(T_0)$.

an instability. In the region where $Z_0(E) < 0$, deviations of the electron distribution function f from equilibrium grow and aggregate instead of decreasing and spreading out [43,44]. Flipping the sign of Z_0^{-1} in Eq. (9) is equivalent to reversing the arrow of time. As a result, the system moves away from rather than toward the equilibrium. Another way to see this is to notice that, according to Eq. (8), $\dot{f} > 0$ at the maxima of $f(E,t)$ [$f' = 0$ and $f'' < 0$] and $\dot{f} < 0$ at the minima when $Z_0 < 0$. Opposite to deviations from stable equilibria, maxima of $f(E,t)$ grow and minima decrease. Straightforward numerical simulations of the full nonlinear kinetic equation (1) confirm this behavior (see Fig. 1), i.e., the system is unstable whenever

$$\min_E Z_0^{-1}(E) < 0. \tag{10}$$

Note also that neglecting the $-2ff'$ term in Eq. (8) and looking for a solution of the form $f \propto e^{-\theta t + ikE}$, we find

$$\gamma(E) \equiv \mathrm{Re}[\theta(E)] = T_0 Z_0(E) k^2. \tag{11}$$

Although this solution is good only at small times and for $E \gg T_0$, it reproduces the main features of the exact solution of the full kinetic equation (1). In particular, we see that the solution grows in regions where $Z_0 < 0$. The growth rate is largest for large k, indicating a highly oscillatory behavior, and diverges at the two points where $Z_0^{-1}(E) = 0$; cf. Fig. 1.

To investigate this instability at finite λ and for various phonon spectra, we perform linear stability analysis of the kinetic equation (1). Linearizing this equation in small deviations from equilibrium and substituting (Sec. 79 of [45])

$$\delta f = f - f_0 = -f_0' \varphi, \tag{12}$$

we obtain

$$A \cdot \dot{\varphi} = -M \cdot \varphi, \tag{13}$$

where $A \cdot \dot{\varphi} = \int d\tilde{E} A_{\tilde{E}E} \dot{\varphi}(\tilde{E})$ and similarly for $M \cdot \varphi$. The metallic state is stable when all separable solutions of Eq. (13) decay exponentially, $\varphi \propto e^{-\gamma t}$ with $\gamma > 0$, i.e., when all eigenvalues γ of the generalized eigenvalue equation

$$\gamma A \cdot \varphi = M \cdot \varphi \tag{14}$$

are positive. Since A and M are real symmetric and M is positively defined [37], γ are real. It is straightforward to see that at weak coupling, $\lambda_k \to 0$, A is positively defined as well [37] and, therefore, all $\gamma > 0$. As we increase the coupling, one of the eigenvalues diverges at $\lambda = \lambda_c$ (when A becomes singular) and becomes negative for $\lambda > \lambda_c$ (Fig. 2). The same behavior is seen in Eq. (11), and λ_c coincides with λ obtained from $\min_E Z_0^{-1}(E) = 0$ up to a relative error of order of 10^{-4} [37]. Note that initial conditions are irrelevant in linear stability analysis; instability arises if a single normal mode is unstable.

It is important to mention that the stability condition (10) underestimates λ_c due to the quasiclassical nature of the kinetic equation (1), which ignores the quantum uncertainty in the electron energy [39]. The length δE of the interval of energies where $Z_0(E) < 0$ needs to be comparable to the energy uncertainty ΔE to ensure the instability. Consider, for example, Einstein phonons, $\alpha^2 F(\omega) = (\lambda\Omega/2)\delta(\omega - \Omega)$, at $T = 0.183g > T_c$. Using Eq. (5), we find that $Z_0(E)$ touches the horizontal axis at $E_{\rm min} = 1.44\Omega$ and $\lambda_c = 1.27$. Since all

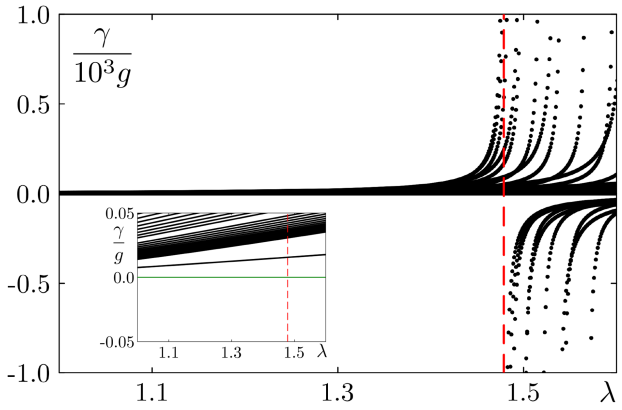

FIG. 2. Results of the linear stability analysis: generalized eigenvalues γ vs the electron-phonon coupling λ at temperature $T_0 = 0.20g$. The metal is unstable when at least one $\gamma < 0$. At this temperature, the instability takes place at $\lambda_c = 1.4788$. The inset shows the part of the spectrum with smallest $|\gamma|$. Note the polelike divergencies in $\gamma(\lambda)$. The first of them is at $\lambda = \lambda_c$ when the inverse quasiparticle weight first touches the horizontal axis, i.e., $\min_E Z_0^{-1}(E) = 0$, see the main text and [37] for more details. Phonon frequencies and couplings λ_k are the same as in Fig. 2 of [37].

relevant energies are of the order of Ω, the uncertainty $\Delta E \sim \Omega$. However, $\delta E \geq \Omega$ only for $\lambda \geq 3.40$. On the other hand, $\delta E > \Delta E$ already at $T = T_c$ in the strong coupling regime [37].

The instability of the metallic state we discovered is an absolute instability (as opposed to metastability). Instabilities of this type are often accompanied by the violation of fundamental thermodynamic inequalities. Indeed, we mentioned earlier that the electronic specific heat turns negative at sufficiently large λ. Even though electrons are strongly coupled to the phonons and the total specific heat is positive [33], the fact that electrons retain many features of an independent system [38] does suggest that $C_{\rm el} > 0$ is required for stability as usual. In particular, the rate of change of the total energy of the electron subsystem and, as a consequence, $C_{\rm el}$ depend only on the electron distribution $f(E,t)$ (and not on the phonon distribution). Moreover, the r.h.s. of the kinetic equation (1) has the same form for any electron-phonon coupling. This suggests that the heat flows from hot to cold as usual. If the electrons are initially hotter and $C_{\rm el} < 0$, their temperature will only increase and the system will never equilibrate.

To see this explicitly, i.e., to prove that $C_{\rm el} < 0$ is a *sufficient* condition for the instability, it is helpful to incorporate the electron-electron collisions into the kinetic equation (1),

$$(1-\Sigma')\dot{f} + f'\dot{\Sigma} = a_{\rm ep} I_{\rm ep} + a_{\rm ee} I_{\rm ee}, \qquad (15)$$

where $I_{\rm ee}$ is the electron-electron collision integral. This corresponds to $M = a_{\rm ep} M^{\rm ep} + a_{\rm ee} M^{\rm ee}$ in the linearized kinetic equation (13), where $M^{\rm ee}$ is the linearized $I_{\rm ee}$. We weighted the collision integrals with positive coefficients $a_{\rm ep}$ and $a_{\rm ee}$ for the sake of the argument; in the actual dynamics $a_{\rm ep} = a_{\rm ee} = 1$. Suppose the metal is stable with respect to the electron-phonon interaction ($a_{\rm ep} = 1$ and $a_{\rm ee} = 0$), i.e., all eigenvalues $\gamma > 0$ when $M = M^{\rm ep}$. Then, Eq. (14) implies that A is positively defined. Since any linear combination $M = a_{\rm ep} M^{\rm ep} + a_{\rm ee} M^{\rm ee}$ of positively defined real-symmetric matrices $M^{\rm ep}$ and $M^{\rm ee}$ [37] with positive coefficients is positively defined and given that A is positively defined as well, it follows that $\gamma > 0$ also for $M = a_{\rm ep} M^{\rm ep} + a_{\rm ee} M^{\rm ee}$ with arbitrary positive $a_{\rm ep}$ and $a_{\rm ee}$. In other words, if the metal is stable with respect to strong electron-phonon interaction, Eq. (15) must also be stable for arbitrary $a_{\rm ep} > 0$ and $a_{\rm ee} > 0$.

Let $a_{\rm ee} \gg a_{\rm ep} > 0$. Then, electron-electron collisions are much faster and, therefore, $f(E,t)$ quickly equilibrates, $f(E,t) = f_0[T(t)]$, at a temperature $T(t)$ corresponding to the instantaneous total energy of the electronic subsystem. To see this, note that $I_{\rm ee}$ vanishes for $f(E,t) = f_0(T)$ for any T and electron-electron collisions conserve the total electron energy density ε. Indeed, multiplying Eq. (1) by E and integrating, we find [37,38]

$$\dot{\varepsilon} = 2\nu_0 \int dE\, E \left[(1-\Sigma')\dot{f} + f'\dot{\Sigma} \right] = 2\nu_0 a_{\rm ep} \int dE\, E I_{\rm ep},$$

since $\int dE\, E I_{\rm ee} = 0$. Here, ν_0 is the density of states at the Fermi level per spin projection. Further, substituting $f(E,t) = f_0[T(t)]$ into the last equation, we obtain [37]

$$C_{\rm el}\dot{T} = 4\nu_0 a_{\rm ep} \int_0^\infty d\omega\, \alpha^2 F(\omega) \omega^2 [N_0(T_0) - N_0(T)]. \qquad (16)$$

By definition, a kinetically stable system must be stable for any small deviation from the equilibrium, so it is sufficient to demonstrate instability for a particular initial condition. Suppose $f(E, t=0) = f_0(T_{\rm in})$ with $T_{\rm in}$ slightly above T_0. Since $\alpha^2 F \geq 0$ and $N_0(T)$ is a monotonically increasing function of T, the r.h.s. of Eq. (16) is negative for any $T > T_0$. If $C_{\rm el} < 0$, $T(t)$ will grow exponentially proving that our assumption that the metal is stable is incorrect.

We emphasize that our proof does *not* assume that electrons *actually* equilibrate among themselves quicker than with the lattice. This does happen under certain circumstances justifying the so-called two-temperature model, where electrons are in quasiequilibrium at their own temperature $T(t)$, which slowly relaxes to the lattice temperature [46–52]. This model is frequently employed in the studies of thermal relaxation of photoexcited metals and operates with equations similar to Eq. (16). In contrast, in our proof Eq. (16) emerges as a consequence of the assumption of stability and does not necessarily describe the actual dynamics. In other words, the statement is that Eq. (16) must be stable provided that the metal is linearly

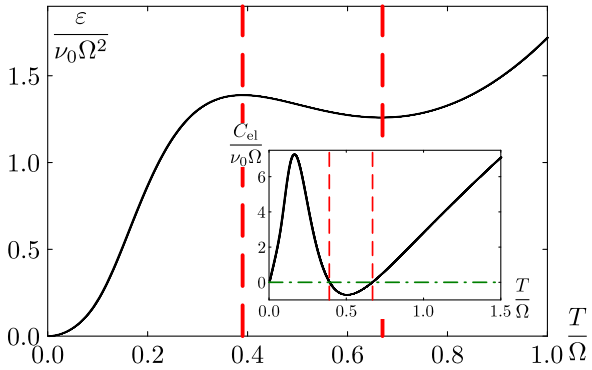

FIG. 3. Electronic internal energy density ε vs (T/Ω) for Einstein phonons with $\lambda = 4.5$ (inset: plot of the electronic specific heat $C_{\rm el}$). The metal is absolutely unstable in the temperature range from $T_- = 0.39\Omega$ to $T_+ = 0.67\Omega$ (vertical dashed red lines). For $T_- < T < T_+$, the slope of the $\varepsilon(T)$ is negative, i.e., $C_{\rm el} < 0$. Cf. isotherms $p(v)$ of a van der Waals gas that similarly feature regions, $v_- < v < v_+$ of absolute instability where the compressibility $\beta = -(\partial p/\partial v) < 0$. The system therefore cannot exist in a purely metallic state for $T_- < T < T_+$ similarly to how the van der Waals gas cannot exist in a purely gaseous or purely liquid state for $v_- < v < v_+$.

stable with respect to the electron-phonon interaction. This is true regardless of whether or not electrons equilibrate among themselves quicker than with the lattice in the course of their actual dynamics. The latter is described by Eq. (15) with $a_{\rm ee} = a_{\rm ep} = 1$ and not Eq. (16). Also, note that we need not worry about the energy uncertainty in this case, since $C_{\rm el}$ is an integral over all energies.

The above instability of metals is analogous to that of van der Waals gases. Recall that below a certain critical temperature an isotherm $p(v)$ of this gas shows a segment $v_- < v < v_+$ with positive slope, i.e., negative compressibility $\beta = -(\partial p/\partial v) < 0$ [53,54]. Similarly, the curve $\varepsilon(T)$—electron internal energy density as a function of temperature—shows a segment $T_- < T < T_+$ with negative slope $[C_{\rm el}(T) < 0]$; see Fig. 3. In both cases, this signals a discontinuous (first order) phase transition, but does not by itself determine the nature of the new phase. From complementary considerations, we know that it is a gas-liquid transition for the gas, and a structural transition to an insulator or another metal with weaker electron-phonon interaction for the metal [33].

In contrast to metastable states, such as an overheated liquid, states with $\beta < 0$ or $C_{\rm el} < 0$ are absolutely unstable, i.e., unstable already to infinitesimal deviations from the equilibrium. Consider a gas with $\beta < 0$ confined to a vertical cylinder by a piston with a load that balances the pressure of the gas [53]. Suppose we slightly decrease the weight of the load. The gas will then expand, but the pressure will grow, and the piston will continue to accelerate upward until it reaches the point $v = v_+$. If we increase the weight of the load, the piston will accelerate downward until $v = v_-$. Similarly, if $C_{\rm el} < 0$ and the electron temperature T is initially slightly above (below) the lattice temperature, it will grow (decrease) until $T = T_+$ $(T = T_-)$.

It is important to bear in mind that the kinetic instability we identify is an inherently nonequilibrium phenomenon, and its proof does not rely on ME theory. The kinetic equation becomes unstable *before* $C_{\rm el}$ becomes negative [55]. In other words, the instability arises before the equilibrium ME theory is able to detect it. Further underscoring the fundamental difference from the P/CDW transition is that the kinetic instability can proceed along entirely different routes. A representative example is provided by hydrides such as H_3S, LaH_{10}, and YH_9, where increasing λ through decompression results in hydrogen diffusion and eventual formation of new compounds rather than a P/CDW transition [56–61]. This process provides a natural experimental setting to probe the onset of the kinetic instability discussed here. Although the initial dynamics of the instability is uniform, spatial structure may emerge later due to secondary instabilities involving couplings across length scales [37]; a detailed analysis of this late-time behavior is beyond the scope of the present work.

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Data availability—The source code required to reproduce figures in this Letter and in [37] is openly available at [62].

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