Removal of the degeneracy in the description of electron-hole–cloud configurations

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The prediction of the stable configuration of an ideal uniformly volume-pumped electron-hole cloud involves the rationalization of a "free-boundary" problem. That is to say, there are an infinite number of degenerate steady-state solutions which satisfy the fluid-dynamical equations and the boundary conditions. If the stationary states are metastable due to isolation or freedom from large-scale fluctuations in the cloud, then all states are attainable via differing initial conditions (e.g., variable risetime of the laser pump). On the other hand, if configurational changes can occur by fluctuation, creation, and annihilation of drops at any "stationary" state, however slowly, then it is necessary to specify a global thermokinetic optimum if a unique stable solution is to be identified. Both the uniquely stable and hysteresis cases are experimentally observed. In this contribution we solve the fluid-dynamical equations for a cloud, explicitly demonstrating the degenerate nature of the boundary-value problem. The global dissipation rate is evaluated and shown to be a function of the exciton-gas density as a unique internal order parameter possessing two zeros and a maximum value, and free of explicit configurational parameters. The hypothesis of a stable *maximal* state in a topologically fluctuating milieu (already suggested by Onsager's dissipation principle) leads near the saturation line to strictly linear global *and* local flux-force relations as Euler-Lagrange equations and to explicit expressions for the drop density and exciton density which are in good functional and fair quantitative accord with conventional, low-excitation stripe and cylinder pumped experiments. Our theoretical expression for the drop radius is less successful, because, we believe, of the inappropriateness of the available experimental test and the strong experimental artifacts.

INTRODUCTION

In this phenomenological treatment we investigate the striking low-temperature, laser-pumped steady-state configuration of electron-hole drops and clouds,[1–6] a phenomenon which possesses the essential elements and complexity of a broad range of self-organizing cooperative systems. We thus see the phenomenon as a paradigm for this nonlinear class of irreversible processes. Noteworthy is the fact that there exists a degeneracy or internal degree of freedom within the steady-state solutions of the field equations for the cloud at the steady state, i.e., an infinite number of spatial configurations satisfy the boundary conditions for ideally uniform volume pumping. If one conceives of this spectrum of states as coupled by macroscopic fluctuations or defect mechanisms, then it is natural to ask whether nature seeks some optimum or stable state within the spectrum. Onsager raised the same question obliquely in relation in turbulence in fluids.[7] The query has also been raised recently by Nicolis and Prigogine[8] and has already been answered positively in relation to analogous model systems which are reviewed in Refs. 9 and 10. In the following we are led to a qualified positive answer for the electron-hole–drop problem. The qualification arises not from a defect in the analysis, but from the fact that the relevant spatial experiments have invariably had ill-defined boundary conditions.

In earlier publications[9,10] it has been argued from the general form of Onsager's dissipation theorem that near saturation the stability point within the internal degree of freedom identified here should lie at a *maximum* in the dissipation rate.[11–13] The application of this principle leads to the required quadratic form for the *global* entropy production, to a linear relation between both the *global* and *local* fluxes and forces as Euler-Lagrange equations, and to a satisfactory closure of the optimal predictions with the experiments in germanium. It is therefore concluded that if a Markoffian macroscopic fluctuation process effecting configuration change is operative within the cloud, then the maximal character of the stability point will have been validated. This, of course, is the basic and successful assumption of classical nucleation theory.

The "free-boundary" problem identified here has been previously encountered and explored in a variety of ways.[3,4,14–16] It only comes into sharp focus, however, within the phenomenological structure presented for the first time in this article.

IDEAL BOUNDARY-VALUE PROBLEM

Laser pumping of pure, defect-free Ge at frequencies in excess of the band gap (~ 0.8 eV) creates an exciton gas which can condense into a cloud of Fermi (electron-hole) liquid drops at temperatures below 6.5 K and, which beyond a certain threshold power, sustains a steady-state configuration. While the decomposition process should strictly be treated within the spinodal formalism,[17] we here proceed in accord with the physics literature, which

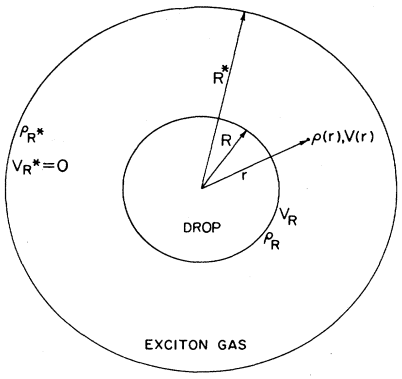

FIG. 1. Boundary values for the ideal electron-hole problem. Note that due to crystallographic effects the drop is almost certainly not a sphere.

follows the classical nucleation and growth formalism.[1-5,16]

For a stationary and uniformly volume-pumped region, the cloud can be approximated as a close-packed lattice, each cell being spherical of the dimensions indicated in Fig. 1. Because the recombination times of both exciton (τ) and electron holes (τ_0) are large (~ 5 and 25 μsec, respectively), a quasiequilibrium phase configuration is created which scales reasonably well to conventional metal-vapor systems with a phase diagram and critical temperature ($\rho \sim 10^{17}/\text{cm}^3$ and $T_c \sim 6.5$ K). The drop surface tension at low temperatures is about 10^{-4} erg/cm^2 which implies critical drop radii $\lesssim 0.1$ times the stable values.[3] At temperatures where the mean-free path is less than the drop radius ($\gtrsim 3$ K), we may represent the exciton-gas phase as a classical fluid of density ρ and local velocity V and solve the steady-state boundary-value problem defined by Fig. 1. This requires the continuity equation for the gas (neglecting viscous dissipation)

$$G-\frac{\rho}{\tau}=\frac{1}{r^2}\frac{\partial}{\partial r}(r^2\rho V)\ , \tag{1}$$

where G is the creation rate of excitons (proportional to pump power) and the isothermal Navier-Stokes (force) equation incorporating the perfect-gas law. Because of the assumed isotropy of the ideal cloud, we are justified with many others in neglecting phonon-wind effects[4,18,19] and writing the force equation with source term originating in (1) as

$$-\frac{\partial\rho}{\partial r}\frac{kT}{m^*}=\rho V\frac{\partial V}{\partial r}-\frac{2\rho V^2}{r}+\left[G-\frac{\rho}{\tau}\right]V\ , \tag{2}$$

where m^* is the effective exciton mass. The steady-state condition in terms of the electron-hole—drop density ρ_0 is

$$4\pi R^2\,|\,V_R\,|\,\rho_R\simeq\frac{4\pi R^3}{3}\frac{\rho_0}{\tau_0}\ . \tag{3}$$

There are two further loose thermokinetic conditions which must be analytically accomodated, viz.,

$$G\tau\geq\rho\geq\rho_{\text{eq}},\quad 0\leq|\,V_R\,|\leq V_T/4\ , \tag{4}$$

where ρ_{eq} is the saturation concentration of excitons and V_T is the thermal velocity. As Fig. 1 shows, there are five unspecified boundary values, ρ_R, ρ_{R^*}, R, R^*, and V_R along with the above four constraints by which they may be determined. Because there is an internal degree of freedom, this representation may be classified with the well-known "free-boundary" problems.[9,10]

Now we may justify *a posteriori* the conventional approximation

$$\rho\cong\rho_{R^*}=\rho_R\ . \tag{5}$$

Hence we can integrate (1) to

$$V=V_R\frac{R^2}{r^2}+\frac{1}{3}\frac{(G\tau-\rho)}{\rho\tau}\left[r-\frac{R^3}{r^2}\right]\ . \tag{6}$$

Evaluating (6) at $r=R^*$, $V=0$ with $R^*\gg R$, yields the volume fraction f via (3) as

$$f=\frac{R^3}{R^{*3}}\cong\frac{G\tau-\rho}{\rho_0}\frac{\tau_0}{\tau}\ . \tag{7}$$

This new result is the kinetic equivalent of the "lever rule" used for determining volume fractions in equilibrium chemical thermodynamics and is quite reasonably augmented by the ratio $\tau_0/\tau>1$. Since (1) applies irrespective of the value of the mean-free path and (2) does not enter explicitly, (7) may be regarded as valid for all temperatures.

The degree of freedom remains, as may be seen by exploration of the special case accommodated by (4), viz.,

$$\rho=\rho_{R^*}=\rho_R=\rho_{\text{eq}}\ , \tag{8}$$

where the saturation ρ_{eq} is a known function of temperature (the phase diagram). In this case we obtain one independent relation in the two unknowns $|\,V_R\,|$ and R, viz.,

$$|\,V_R\,|\rho_{\text{eq}}=\tfrac{1}{3}R\frac{\rho_0}{\tau_0}\ . \tag{9}$$

We now proceed to calculate the excess free-energy dissipation rate for the continuum model. In principle it is possible to proceed via the quantum-statistical methods of Klein and Meijer[20] and Chambers.[21] However, for conciseness and consistency we proceed here via a chemical method which attributes the dissipation to a chemical condensation reaction. That is to say, we evaluate the isothermal dissipation of free energy as the integral flux across the surface per unit of cloud volume times the free-energy change per exciton.

$$T\sigma=|\,V_R\,|\rho\,\Delta F\frac{4\pi R^2}{\frac{4}{3}\pi R^{*3}}\ , \tag{10}$$

where ΔF is taken as the ideal-gas expression

$$\Delta F=kT\ln\frac{\rho}{\rho_{eq}}\ . \tag{11}$$

This neglects loss of coherency of the outgoing radiation. Introducing (3) and (7) and manipulating, yields

$$T\sigma=\frac{G\tau-\rho}{\tau}kT\ln\frac{\rho}{\rho_{eq}}=JX\ , \tag{12}$$

the latter expression defining the global flux and force conventionally as a mass rate and a free-energy difference. In this we are modeling the reaction such that the excitons are bled at mean velocity $|V_R|$ into an infinitesimal reservoir of density ρ_{eq} at the surface and are instantly absorbed nondissipatively at their equilibrium density. It is interesting to note that the form of (12) is similar to that for a two-level quantum-statistical system which is decoupled from the radiation field.[20,21] Furthermore, it is significant that the configurational parameters do not appear explicitly.

Now ρ in (12) is an internal order parameter, surrogate for internal configurational and surface changes allowed by the degree of freedom identified earlier. We now suppose, in accord with our earlier discussion, that the stability point is at a maximum of $T\sigma$ with respect to ρ. Approximating (12) near saturation by

$$T\sigma=\frac{kT}{\tau\rho_{eq}}(G\tau-\rho)(\rho-\rho_{eq})\ , \tag{13}$$

we find a stable value of

$$\rho=\frac{G\tau+\rho_{eq}}{2}\ , \tag{14}$$

i.e., the optimum is at a mean of the two possible extremes. Furthermore, the optimum value of ρ implies

$$T\sigma=\frac{kT}{4\tau\rho_{eq}}(G\tau-\rho_{eq})^2\ , \tag{15}$$

and

$$J=\frac{\rho_{eq}}{\tau kT}X\ , \tag{16}$$

which is to be regarded as the linear Euler-Lagrange equation for the maximal dissipation principle.

It may be noted now that (12) has two zeros. The conventional (equilibrium) zero lies at the phase boundary and is defined by

$$G\tau=\rho=\rho_{eq}\ . \tag{17}$$

There is a second kinetic zero which can be identified when R, by a virtual excursion defined by (7) at fixed $G\tau$, reaches such a large magnitude that ρ is pulled down to ρ_{eq} by drop recombination (refer to Fig. 1). Via (7) and (3) this can be represented as a conditional maximum $|V_R|$ as well as a maximum R virtual state of zero dissipation. It is a superconducting state in which the functional $J\rightarrow$ max as $X\rightarrow 0$ [cf. Eq. (13)].

It remains to establish the unknown function V_R. As previously, we will ignore capillarity (i.e., the possible R dependence in V_R). Now from (10) and (12) we can write for the global flux

$$J=G-\frac{\rho}{\tau}=|V_R|\rho\frac{4\pi R^2}{\frac{4}{3}\pi R^{*3}}=|V_R|\rho B\ . \tag{18}$$

As expected on general grounds, this states that the rate of survival of injected excitons per unit volume is proportional to the local flux across a drop boundary, the proportionality factor being the drop area per unit volume (B). Now we recall that the variational principle acting on (12) or (13) does not involve the geometric factor explicitly. This means that the global variation can follow an infinity of geometric paths. It is convenient to construct the variational path for $|V_R|$ within this spectrum such that the area per unit volume

$$B=B(G)\ , \tag{19}$$

since this assures that the global and local fluxes are strictly proportional at fixed pump power, and since J is linear with the force at the optimum, this will also be the case for the local flux $|V_R|\rho$. The optimal principle may thus be associated with a local flux-force relation in accord with the principles of near-saturation irreversible thermodynamics.

Now we can identify the maximum possible velocity as equal to the cosine law averaged thermal velocity $V_T/4$ where

$$V_T=(8kT/\pi m^*)^{1/2}\ , \tag{20}$$

which, in view of our earlier discussion and reference to Fig. 1, may only be attained via conditional maximum states in $|V_R|$ corresponding to $\rho=\rho_{eq}$. Consider such a state with reference to Fig. 1 and Eq. (3) subjected to a continuous increase in pump power at R^* fixed so that R and thus $|V_R|$ must continuously increase until the maximum possible value is reached with $G\gg\rho_{eq}$. Thus we asymptotically establish

$$B=\frac{4G}{V_T\rho_{eq}}\ . \tag{21}$$

Since B is independent of ρ, we may write, in general, that $|V_R|$ follows a virtual path in ρ, consistent with (3) and (18), such that

$$|V_R|=\frac{V_T}{4}\frac{\rho_{eq}}{\rho}\frac{G\tau-\rho}{G\tau}\ , \tag{22}$$

which at the near-saturation optimum takes the value

$$|V_R|=\frac{V_T}{4}\frac{\rho_{eq}}{G\tau+\rho_{eq}}\frac{G\tau-\rho_{eq}}{G\tau}\ . \tag{23}$$

We can now define the local force as

$$X'=kT\frac{G\tau-\rho_{eq}}{G\tau}\ , \tag{24}$$

so that

$$J'=\frac{V_T\rho_{eq}}{8kT}X'\ . \tag{25}$$

Note that the variational path which may be associated with (25) [within the general spectrum allowed by (12)] corresponds to a fixed areal density ($B=$const), a constraint which is now seen to be essential.

Note also that $J'X'$ represents the dissipation per unit area as required, whereas the global JX is per unit volume. We conclude that the variational principle has generated

both a global and a local isothermal invariant in the respective ratios of fluxes to forces [cf. Eqs. (16) and (25)], a result of considerable significance to the general theory of free boundaries. Indeed, the delicate self-consistency requirements revealed here are the very essence of cooperative phenomena.

It may seem unusual at first sight that the quasiequilibrium quantity ρ_{eq} dominates both the global and local rate constants. On the other hand, we can approximate ρ_{eq} empirically by

$$\rho_{eq}/\rho_0 \cong \exp(-\phi/kT) , \tag{26}$$

where $\rho_0 \cong 2\times 10^{17}/\text{cm}^3$ (Ref. 22) and $\phi \cong 2.4$ MeV.[16] Since ϕ is close to the value of the binding energy, this carries essentially the same temperature dependence as the Dushman equation for evaporation. Since near equilibrium the forward and backward rate constants are proportional (detailed balance), Eq. (26) may be expected to represent the forward reaction as well. Interpreting ρ_0 as the drop-level occupancy, (26) may also be interpreted as a Boltzmann factor from which the ideal work of formation of the drop by gas compression is $\phi = kT \ln \rho_0/\rho_{eq}$, thus verifying that the activation barrier and its temperature dependence for the forward and backward reactions are equal.

To finally validate the approximation leading to (5) and (6), we note from (23) that the stable $|V_R| \leq 0.043 V_T$ for all $G\tau/\rho_{eq} > 1$. Manipulating (6), (7), and (2) at the interface, we find that $\partial\rho/\partial r$ will, as a result, be vanishingly small as required for approximation (5).

The five formulas (3), (5), (7), (14), and (23) now represent a complete, near-saturation solution for the five independent variables. This result was effected by the use of the maximal principle to remove the degree of freedom identified. The predictions are in functional and order-

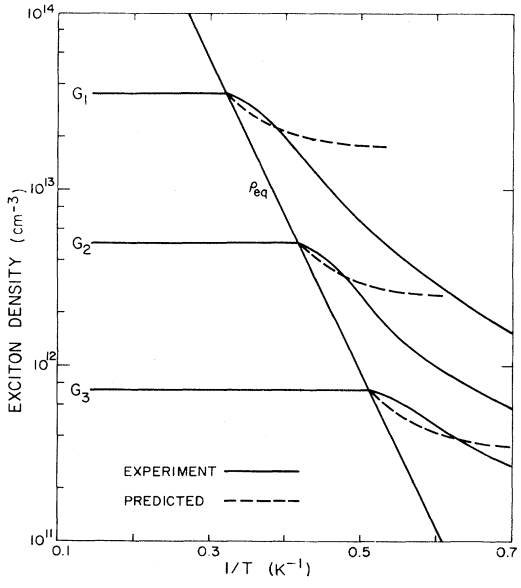

FIG. 2. Exciton density within cloud as function of temperature and pump power.

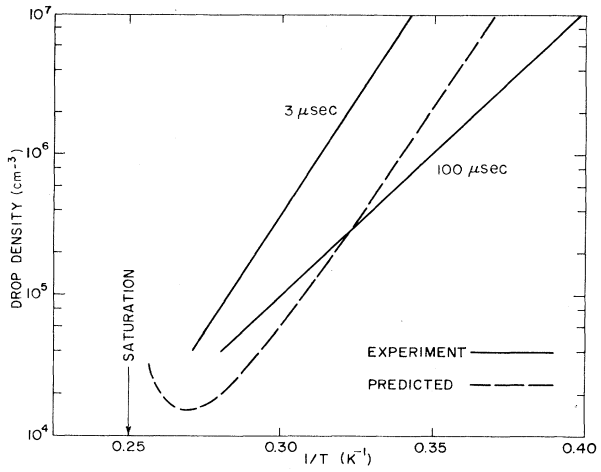

FIG. 3. Drop density as a function of $1/T$. Hysteresis effects associated with risetime are evident in the experiments (see text).

of-magnitude accord with most of the relevant experiments. The directly testable relations are (14), the steady-state radius

$$R = \frac{3V_T\tau_0\rho_{eq}}{8\rho_0}\frac{(G\tau-\rho_{eq})}{G\tau} , \tag{27}$$

and the density of drops

$$N_d \cong \frac{1}{\frac{4}{3}\pi R^{*3}} = \frac{f}{\frac{4}{3}\pi R^3} = \frac{64 G\rho_{eq}^{-3}}{9\pi V_T^3}\left[\frac{\rho_0}{\tau_0}\right]^2 \frac{1}{(1-\rho_{eq}/G\tau)^2} . \tag{28}$$

In the latter transformation, the optimum

$$f \cong \frac{1}{2}\frac{(G\tau-\rho_{eq})}{\rho_0}\frac{\tau_0}{\tau} , \tag{29}$$

has been incorporated. It is particularly to be noted that because $\rho \cong$ const, the force balance does not enter explicitly into the testable results. Consequently, the latter are independent of our omission of a phonon-wind term in the Navier-Stokes equation (2), depending only on the mass balance and the entropy production principle.

EXPERIMENTAL CLOSURE

Experiments have yet to be carried out in the ideal, uniform volume-pumped configuration. Consequently, we focus on the experiments which best approximate to this ideal, those which employ cylindrical and stripe geometry.

Let us now represent the best-fit germanium exciton phase boundary by (26), with $\rho_0 = 7\times 10^{16}$ and $\phi = 2.2$ MeV,[16,23] and consider the comparisons with experiment given in Figs. 2–4. In these experiments an approach to uniform volume excitation was achieved in a plane or cylinder of about 0.2-mm crossdimension. The exciton density measurements of Fig. 2 were calculated from the measured absorption and photoconductivity spectra. The absolute value of the exciton concentration at a given excitation level was evaluated from the absorption coefficient

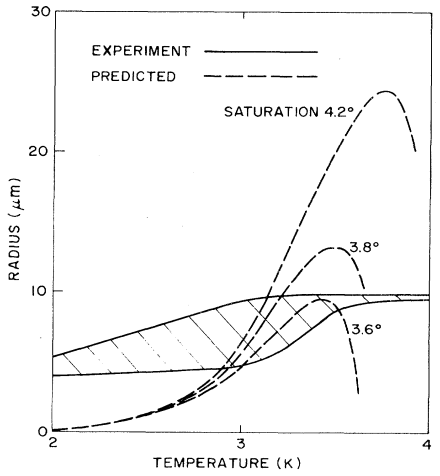

FIG. 4. Drop radius as a function of temperature and pump power. Deviations between theory and experiment are due in part to experimental artifacts.

in photoionization of the exciton.[23] The drop density and radii measurements of Figs. 3 and 4 were obtained through Rayleigh scattering and absorption of 3.39μ radiation from the cloud.[24] In the latter experiments, results for laser pump rise times of ~ 1 μsec and 100 μsec were obtained. For the theoretical calculations, representative values of $m^* \sim 0.6\times 10^{-27}$ g, $\tau_0 \sim 25$ μsec, $\tau \sim 5$ μsec, and $\rho_0 \sim 2.5\times 10^{17}/\text{cm}^2$ (Refs. 3, 4, and 22) were used.

Since the threshold line of Gershenzon *et al.* is very close to Westervelt's phase diagram ($\rho=\rho_{eq}$) we have assumed it to be identical and have plotted our near-saturation prediction from (14) on an abridgment of Gershenzon's diagram (Fig. 2). The agreement near saturation is fair. The disagreement further from saturation is due in part to our theoretical approximation [linearization of Eq. (12)], but more significantly to the necessary expansion of the experimental drop and gas cloud via the phonon wind at the higher supersaturations.

Figure 3, after Bagaev *et al.*,[24] demonstrates fair quantitative agreement for the prediction of drop density N_d as a function of temperature near saturation. The other density observations of Bagaev *et al.* for the same pump power (not shown) tend to saturate at $1/T > 0.4$ due apparently to heating effects. Figure 3 also indicates a hysteresis effect associated with risetime which we will discuss later.

The agreement between theory and experiment for the drop radii is the least satisfactory. This we believe is due to experimental artifacts and complex hysteresis effects. The hatched experimental band in Fig. 4 represents a range of pump powers corresponding to saturation temperatures $T_0 \sim 3.6$—4.2 K and rise times of 1 and 100 μsec. We conjecture that the observed low-temperature excess radii (together with the saturation of N_d) is due to overheating of the high-density cloud associated with sharply increased dissipation following decreasing temperature at fixed pump power [see relation (15) and Refs. 22 and 24]. That is to say, the actual temperature does not change very much with change in cryostat temperature for the particular strong pump conditions in the low-temperature experiments. Zarate and Timusk,[22] who took great care to avoid temperature effects, have observed low magnitude radii near 2° (Fig. 4).

The strong saturation of the radii at high temperature (Fig. 4) has evoked considerable discussion in the past. This is logically related to our underprediction of N_d via Eq. (28) (cf. Fig. 3). The effect could be attributed to a rather lengthy time of new drop growth in relation to the rate of cloud expansion and associated ejection to the saturation line by the phonon wind.[4,19] Note that no great significance is to be attributed to the reversals in the theoretical N_d and R curves in the very close approach to equilibrium, for here the classical quasiequilibrium thermodynamic model fails. A more rigorous spinodal decomposition model avoids this anomoly as demonstrated by Kirkaldy *et al.*[17,25]

EFFECTS OF DROP PINNING

Martin has demonstrated that doping a germanium crystal inhibits the expansion of the cloud[26] This implies that crystal defects can pin the drops, presumably by electrical interactions. This kind of observation raises the possibility of hysteresis effects, phenomena in fact recorded by Bagaev *et al.* (Fig. 3) and Westervelt.[27] Strong pinning also raises the possibility that after cloud initiation the concentration in the pumped region can drop below the critical value for nucleation and a pinned quasistable array evolve which "remembers" the nucleation history as determined by the rise time. Such an array, by its nature, resists an evolution toward a kinetic optimum. The results of Bagaev *et al.* in Fig. 3 reflect this outcome. We may suppose that more surviving nuclei are generated with a short rise time leading to a higher metastable density N_d (Fig. 3).

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