Homogeneous Nucleation of Vapor Condensation. I. Thermodynamic Aspects*

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In absence of all foreign materials or wall surfaces, phase transitions of the type vapor-to-liquid or liquid-to-solid are blocked by an activation-free-energy barrier. The latter arises from surface-free energy increases resulting from appearance of embryos of the more condensed phase. The thermodynamics of this type of phase transition are examined in Part I for the particular case of vapor condensation, a case for which theory and experiment now stand in tolerably good agreement. In Part II, kinetic aspects will be considered.

1. INTRODUCTION

IT is probably rather widely believed that if air containing water vapor, say on a humid summer day, is cooled to its dew point, droplets of water are certain to form by condensation. Similarly, it is fairly generally thought that if water is cooled to 0°C, freezing of ice is assured. Now although common experience does support both views, these views are incorrect if taken as generalizations applicable under *all* conditions. They become incorrect in those situations where vapors or liquids are cooled under conditions of uncommon purity, characterized by complete absence of foreign materials or wall surfaces which can render subtly important assistance to molecules seeking to condense or to solidify in well-behaved manner. Given completely pure vapor or liquid free from such extraneous influences we find that substances behave under cooling as if condensation point and freezing point simply did not exist—i. e., the pure phases exhibit, respectively, the phenomena of *supersaturation* and *supercooling*. The latter phenomena play important roles in many industrial processes and crop up in a wide variety of problems of basic scientific significance.

From both a theoretical and a practical point of view, one's insight into the mechanism of the everyday mode of "well-behaved" condensation and freezing is greatly enhanced by an understanding of the thermodynamic and kinetic principles of the "anomalies" of supersaturation and supercooling. Indeed, once one appreciates the latter principles, he clearly recognizes that the truly anomalous case is really that of the familiar behavior of condensation at the vapor's dew point or solidification at the liquid's freezing point!

It is the objective of the following discussion to present a summary of the general principles of nucleation of phase transition, using vapor condensation as the example. Because the kinetic theory of gases is well developed, whereas the kinetic theory of liquids is not, it becomes possible to describe rather more thoroughly the nucleation mechanism for condensation of vapor than for solidification from the melt. However, the general principles are quite similar in these two important types of phase transition, so the implications will be broad. In Part I of this discussion, chief emphasis is placed upon *thermodynamic* aspects. Part II will subsequently summarize the main features of the ***kinetics*** of nucleation processes. Emphasis throughout will be placed upon physical interpretation of the basic mathematical description of nucleation theory.

As a point of terminology, it may be noted that the type of nucleation to be considered here is referred to as *homogeneous nucleation* because it takes place in a completely homogeneous phase with no foreign bodies (ions, hygroscopic condensation nuclei, wall surfaces) present. When any of the latter are present and act to assist the system to get over the activation barrier which we shall see is imposed by surface-free-energy considerations, the process is properly called "heterogeneous nucleation." Systems of the inevitable degree of contamination accompanying almost all but scrupulously arranged laboratory experiments afford only examples of *heterogeneous nucleation*. It is our excessive familiarity with the superficial features of such ordinary heterogeneous nucleation processes that

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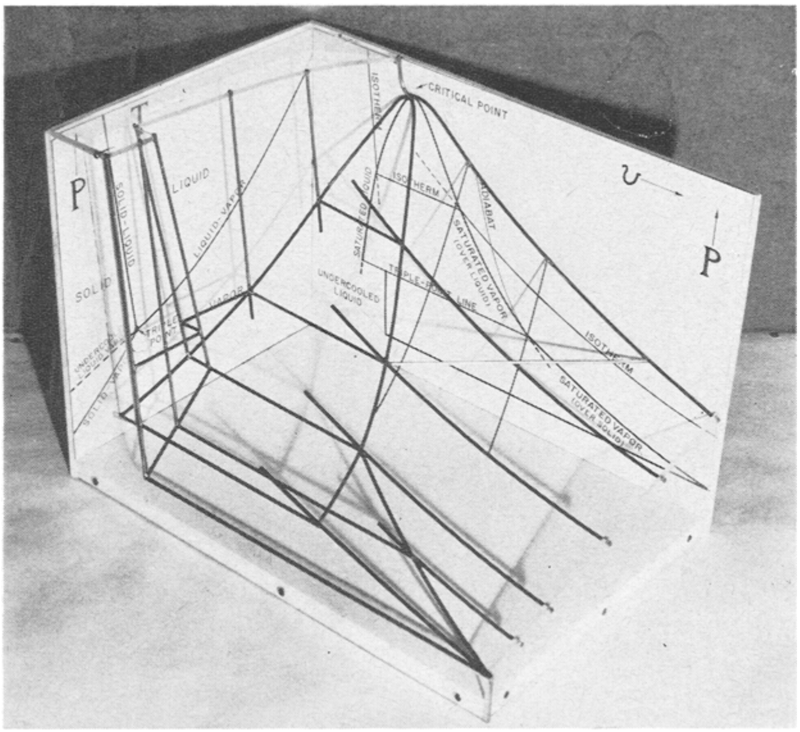

FIG. 1. Wire model of p-v-T surface for a substance such as water. Note metastable extensions of all ideal-vapor isotherms. In absence of heterogeneous nucleation, the system's characteristic point moves smoothly past the saturated vapor locus up along these metastable extensions as pressure is raised isothermally, starting from an ideal vapor state.

stands in the way of clear recognition of the nature of the conceptually more simple and more fundamental homogeneous nucleation processes which will be our main concern in the remainder of this discussion.

The ground work of the theory of nucleation of phase transition might be said to have been laid by Gibbs but the theory was not put into any very specific form until about the third and fourth decades of the present century, notably by Becker and Doering, Farkas, Volmer, Zeldovitch, and others. A fairly detailed, but not uniformly clear treatment of principles is presented by Frenkel.[1] Mason[2] presents almost verbatim a recent modification of Zeldovitch's analysis due to Farley, and gives a good critical discussion of the present state of experimental verification of the theory. A recent publication of the Faraday Society[3] should provide the interested reader with extensive bibliographical references plus several excellent summaries by such workers as Dunning, Mason, Fletcher, and others.

2. FREE ENERGY OF EMBRYO FORMATION

We may bring vapor to a supersaturated state of given temperature T and vapor pressure p (where $p>p'$, p' being the saturation vapor pressure of the substance in question at T) along a variety of paths, of which an isothermal compression path represents one very simple example. As we compress a vapor isothermally, we eventually reach its nominal condensation pressure, i.e., the saturation vapor pressure $p'(T)$ of the liquid phase of the same substance. Elementary discussions typically assert that further attempts to compress the system lead only to formation of steadily increasing amounts of the liquid phase, the characteristic point of the system merely sliding along an isobar-isotherm on the liquid-vapor region of the substance's equilibrium surface. This elementary viewpoint sidesteps the nucleation problem and can be considered correct only if foreign nuclei or wettable walls are assumed present to induce condensation at p'. In the absence of those latter heterogeneous nucleating agencies, the character-

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[1] J. Frenkel, *Kinetic Theory of Liquids* (Dover Publications, New York, 1955), pp. 366 ff.

[2] B. J. Mason, *Physics of Clouds* (Oxford University Press, New York, 1957), pp. 2–11.

[3] Faraday Society, *The Physical Chemistry of Aerosols* (Aberdeen University Press, Faraday Society Discussions No. 30, Aberdeen, 1960), pp. 9–45.

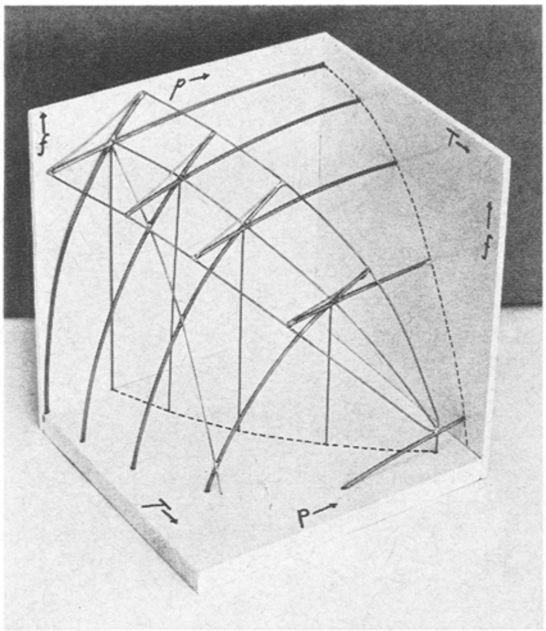

FIG. 2. Wire model of the Gibbs free-energy surface (f-p-T surface) for a pure substance, showing metastable extensions of isotherms and unstable segments of complete van der Waal's isotherms on upper leaf.

istic point of the system merely continues to move up along the *metastable extension* of the ideal-vapor isotherm on which it approached point p', T.

This is illustrated on a three-dimensional wire model of an equilibrium p-v-T surface, designed by the writer for use in classroom discussions of nucleation thermodynamics and shown in Fig. 1. A few representative wire isotherms are shown in the vapor region, and short extensions into the supersaturated vapor region can be seen to represent the metastable supersaturated states attained by isothermal compressions. (This model was designed to illustrate a number of other important thermodynamic principles and processes beyond the scope of this discussion. The writer would point out that such wire models prove more considerably useful in classroom use than molded surfaces of the type commercially available.)

Now as our system's characteristic point moves up the metastable extension of its ideal-vapor isotherm, its specific Gibbs free-energy f (dimensions ergs g^{-1}) rises above that for the liquid phase at the same temperature T. The differential relation $df = vdp - sdT$, where v and s are, respectively, the specific volume and specific entropy of the vapor, may be integrated along the metastable isotherm from saturation pressure p' to some supersaturated pressure p, giving as the *excess* of f over that for liquid at the same temperature T familiar expression

$$\Delta f = RT \ln(p/p'), \qquad (1)$$

where R is the gas constant per gram of the vapor in question. We assume p to be well below the substance's critical pressure, so that the ideal gas law holds. It must be stressed that at the moment we assume *no* appearance of embryonic droplets in our supersaturated vapor. As we shall see below, this is an unrealistic assumption but one that is conceptually useful for the moment.

For any pure substance, it is possible to construct a three-dimensional f-p-T surface. Enlightening discussion of the general form and principal properties of such a surface have been given by Callen,[4] and by Pippard,[5] and from their treatments the writer drew the idea of constructing the wire model shown in Fig. 2. No numbers are entered on the axes since this model is only a qualitative representation. It shows vapor-region isotherms rising steeply (large specific volume) to the locus of intersection with the liquid-region isotherms which have much lower slope, in accordance with the derivative properties of the relation $df = vdp - sdT$. The model suggests the greater *negative* slope (large specific entropy) of the isobars in the vapor as contrasted with the liquid region. But most significantly for the present discussion, the model shows the extensions of all the vapor isotherms that overlie the liquid region near the locus of saturation. (The latter locus is the midrib of the leaf-shaped "surface" extending diagonally across the top of the model. Note that this skew curve has as its projection on the p-T plane the familiar saturation vapor pressure curve, shown in the model as a dotted curve. The verticals are merely supports, though they aid in revealing the preceding projective relationship.) That these extensions, counterparts of those in the p-v-T model of Fig. 1, *overlie* the

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[4] H. B. Callen, *Thermodynamics* (John Wiley & Sons, Inc., New York, 1960), pp. 146–163.

[5] A. B. Pippard, *Classical Thermodynamics* (Cambridge University Press, New York, 1960), pp. 116–121.

liquid region, is consistent with the metastable nature of the associated supersaturated states, for the vapor there has a finite excess of free-energy over that of the liquid at the same temperature. (The cuspate junctions of these metastable extensions and the crossribs on the upper part of the model in Fig. 2 are related to the van der Waals' equation of state, but will not be discussed here. Their significance is best appreciated by consulting Callen's or Pippard's treatments of this type of surface.)

Since it is the spontaneous tendency of thermodynamic systems to assume a state of lowest possible Gibbs free-energy, we must now ask why a system whose characteristic point is somewhere on the metastable supersaturated vapor portions of the surfaces modeled in Fig. 1 and Fig. 2 does not simply jump to the available state of lower f in which newly formed liquid phase is present in equilibrium with the residual vapor. The answer is this: The only possible route to the state of smaller f is *via dropwise condensation* (assuming no foreign nucleants and remoteness of containing walls or else completely nonwettable walls); and as soon as embryonic droplets enter the picture we must reckon with the *surface-free-energy* of the interfaces between the drops and the surrounding vapor. When a bit of vapor condenses, the *decrease* in free-energy due to condensation (bulk-free-energy change of phase transition) varies as the mass condensed, i.e., as the *volume* of liquid droplet formed, while the surface-free-energy *increase* varies as the *area* of the droplet. If r is the droplet radius, the positive term varies as r^2, the opposing negative term as r^3, and regardless of the magnitudes of the coefficients of these terms, the positive r^2 term is mathematically certain to dominate over the r^3 term in *some* sufficiently small r interval above zero. Therein lies the obstacle to attaining the stable state of lower f depicted in Fig. 2. That preferred state lies a finite distance away and may only be reached by a path that crosses the "forbidden region" in which the *increases* in f due to appearance of new surface energy dominate over bulk-free-energy *decreases* in f.

We can describe this mathematically by now taking as a more convenient f-reference state that in which we initially have supersaturated vapor at some given high vapor pressure $p\,(p>p')$ at temperature T, and in which it is assumed that not a single embryonic droplet exists. The latter proviso, it must be carefully noted, is to be construed as ruling out dimers, trimers, and all other clusters of molecules, however small. Then the rise ΔF in the free-energy of our system as we isothermally and (essentially) isobarically form a spherical embryo of some small radius r can be written as

$$\Delta F=4\pi r^2\sigma-(4/3)\pi r^3\rho RT\ln S. \qquad (2)$$

The first term on the right is the intrinsically positive contribution of the surface-free energy, where σ is the specific surface-free-energy having dimensions ergs cm^{-2}. (In liquids, σ is also the familiar surface tension; in the case of the solid-liquid interfaces arising in nucleation from the melt "surface tension" loses meaning, but remains well-defined as the specific surface-free-energy.) The second term, in which ρ is the liquid density and $S=p/p'$ is the vapor supersaturation ratio, represents the contribution to ΔF made by the bulk-free-energy change.[6] Its form is taken from (1), but the sign is reversed, of course, because of our new choice of reference state. The bulk-free-energy change in going from vapor at p', T to liquid inside the drop formally comprises *three* parts. The *first* is that obtained in isothermally lowering the vapor pressure from p to p'. The *second* is obtained in condensing vapor at p', T to liquid at p', T, and is *zero* by the equilibrium property of f. The *third* is obtained in isothermally compressing the *liquid* from pressure p' to the high internal pressure of the liquid within the droplet of radius r, but can be shown to be a numerically quite small contribution in view of the small specific volume of the liquid. (There are alternative ways of formulating the over-all problem here which ignore surface-free-energies as such and instead deal explicitly with the effects of high internal pressure on the free-energy of the droplet. These can be shown to be fully equivalent albeit seemingly dissimilar thermodynamic routes to the same goal.[7]) Of these three contributions, only the first is important and is written as the right-hand member of (2).

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[6] The reader should note carefully that S will, throughout this paper, denote the vapor supersaturation ratio p/p', and not the entropy, for which the same symbol is often used.

[7] J. Frenkel, see reference 1, p. 369.

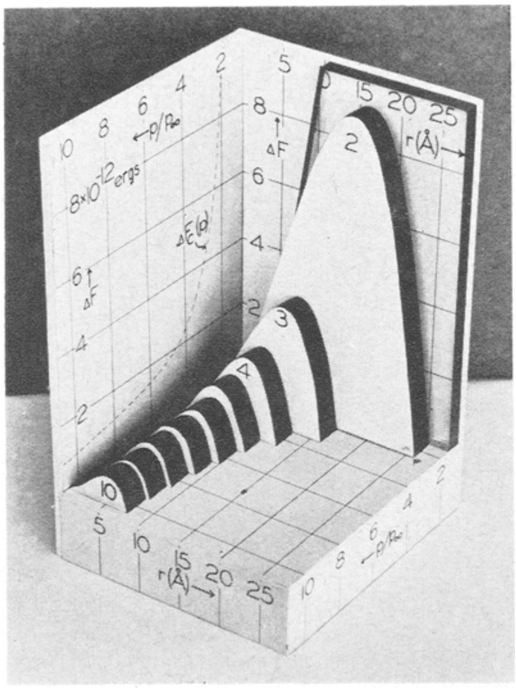

FIG. 3. Wood and plastic model of ΔF-S-r surface for water vapor at 0°C.

Equation (2) specifies ΔF as a function of r and S for fixed T, whence it follows that one may construct a ΔF surface over the r-S plane. A *quantitatively* accurate model of such a ΔF-r-S surface is shown in Fig. 3, calculated for the case of water vapor at 0°C. It displays many thermodynamic features of importance in homogeneous nucleation theory. Note from (2) that for the limiting case $S=1$, the bulk term vanishes and ΔF rises monotonically as a parabola, depicted in the model of Fig. 3 by the transparent plastic lamina in the $S=1$ plane (p' is identified on the model as p_∞, denoting infinite radius of curvature of the liquid surface, the condition tacitly assumed to hold in the customary definition of saturation vapor pressure p'). For *subsaturated* vapors, the ΔF curve would rise even more steeply, because a *fractional* S makes the second term on the right go *positive*, reinforcing the effect due to the surface-free-energy barrier, a thermodynamically quite understandable situation.

For $S>1$, however, the second term is a *negative* contribution to ΔF, its magnitude rising as the degree of supersaturation is raised. Its presence assures existence of a maximum in ΔF at some r, the value of the latter falling as S rises. This very important feature of (2) is depicted in the model of Fig. 3 by the calculated profiles of the wood laminae inserted for integrally rising values of S ranging from 2 up to 10. As we shall note in the next section, *the presence of these maxima is fundamentally significant in nucleation theory.* They represent the summits of the free-energy activation barriers blocking the formation of the new phase, and can only be surmounted as a result of rare fluctuation phenomena in the supersaturated vapor.

It is illuminating to consider somewhat further the cases where $S\leq 1$. Even though such thermodynamically stable cases do not enter directly into phase transition nucleation processes, some of their characteristics shed important light on our problem. Contrary to the ordinary point of view, subsaturated or just-saturated vapor is not completely free of embryos of the liquid phase (nor is supercooled liquid completely free of embryos of the solid phase). Rather, there exists a *statistically steady-state population of embryos* characterized by the Boltzmann-like distribution function

$$n_g = n_1 e^{-\Delta F/kT}, \qquad (3)$$

where now we use the number of molecules g per embryo rather than r as the size parameter of the embryo; n_g is the number per unit volume of vapor of embryos of size g, k being the Boltzmann constant.[8] Here ΔF is, by (2), a rapidly increasing function of r (and hence also of g) because the second term on the right in (2) is nonnegative when $S\leq 1$. Illustrative values of n_g computed from the distribution Eq. (3) for water vapor at 0°C with saturation ratios $S=0.5$ and $S=1$ and for three values of g are given in Table I. One cubic centimeter of half-saturated

TABLE I. Values of n_g (cm^{-3}) for two saturation ratios S.

$g\rightarrow$	5	10	20
$S=0.5$	1×10^{4}	5×10^{-5}	6×10^{-18}
$S=1.0$	3×10^{5}	5×10^{-2}	6×10^{-12}

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[8] J. Frenkel, see reference 1, p. 380, gives a lengthy proof of Eq. (3); the present writer knows of no simple proof for existence of such a Boltzmann-type distribution of embryos. To express ΔF in terms of g and not r, one uses the relation $(4/3)\pi r^3\rho = gm$, where ρ is the density of the liquid phase and m is the mass of one molecule.

water vapor at 0°C is seen to contain the surprisingly large number of ten thousand 5-mers at any instant, some destined momentarily to fly apart, others to be joined by a third to contribute in the next moment to the instantaneous 6-mer population. The concentration of clusters of larger g falls sharply according to (3), and for $S=0.5$ and $g=20$ is already down to about 10^{-17} cm^{-3}. For just-saturated vapor all the n_g are larger, of course, and Table I shows that even 10-mers, embryos that have been built up by a fortuitous concatenation of collisions to an aggregate of ten water molecules, are present at $S=1$ in mean concentration as high as 0.05 cm^{-3}, i.e., an average density of about 1 in every 20 cm^3 of saturated vapor.

However, 10-mers lie well on the low side of the maxima of the ΔF curves (Fig. 3) even for moderate degrees of supersaturation, so their fluctuatory presence in just-saturated vapor by no means provides the necessary path for the system to begin its condensational phase transition to stabilize itself in the face of further compression. Equation (3) predicts still more infrequent occurrence of embryos of size beyond the limit of 20 displayed in Table I. Thus, one finds that a cube of just-saturated vapor some 2.3 miles on edge would, on the average, contain just *one* embryo of size $g=25$, and Eq. (3) shows that one would not find a single cluster of size $g=50$ at a randomly chosen instant unless he had a cube of vapor ten million miles on edge! Trading time for volume, one thus senses that with vapor samples of moderate size (say, cloud-chamber size), he would have to wait for centuries on end before witnessing the chance build-up of an evanescent aggregate containing even only a few tens of molecules in his sample if $S=1$. This is another way of viewing the practically insuperable barrier to phase transition at $S=1$ in absence of heterogeneous nucleants such as ions or wettable surfaces.

In computing values of n_g for Table I, a tacit assumption conventionally made in nucleation theory has been employed, namely that we may proceed without too serious error, to assume that the bulk value of σ continues to have meaning even down to quite small molecular aggregates. This assumption has been discussed from a variety of viewpoints in recent years, summaries of which will be found, for example, in Davies and Rideal.[9] Briefly, weight of argument indicates that we may use bulk values of σ with fair accuracy down to values of g of order 10–20 but below that the effective value of σ probably diminishes and we should resort to some direct molecular-bond calculation. This limitation must be kept in mind in assessing all results for very small g. The above-estimated 5-mer concentration, for example, must be regarded as good to order of magnitude at best.

3. CRITICAL EMBRYOS AND THE KELVIN EQUATION

In discussing Eq. (2) and the model shown in Fig. 3, we have noted that ΔF exhibits a maximum for any case for which $S>1$. Invoking the basic rule that any process in which *F decreases* is a spontaneous process, we see that if, by any means, our system can get up to the summit of its particular ΔF curve, the slightest additional condensational growth of the droplet beyond the size associated with that summit should lead to irreversible growth of the drop at the expense of the supersaturated vapor, and the characteristic point will plunge rapidly down the right-hand portion of the ΔF-r curve (Fig. 3). That is, the r for which ΔF attains its maximum will be a *critical embryo radius* for whatever S we are dealing with.

To determine this critical radius r^*, we differentiate both sides of (2) partially with respect to r holding T and S constant (remembering that while the droplet is as tiny as a subcritical embryo in, say, a cubic centimeter of vapor, its growth will not deplete the vapor stock by an amount producing measurable change in p, nor will its latent heat-release sensibly alter T). On setting the derivative equal to zero and solving for the thereby specified critical radius we find

$$r^*=2\sigma/\rho RT(\ln S). \qquad (4)$$

This relation reveals the very important rule that *the larger the supersaturation ratio S, the smaller the critical radius r**. In Table II the sizes of the critical embryos corresponding to several values of $S>1$ are indicated, expressed in terms of numbers of molecules as well as in

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[9] J. T. Davies and E. K. Rideal, *Interfacial Phenomena* (Academic Press Inc., New York, 1961), p. 11.

TABLE II. Radius r^* and number of molecules g^* per critical embryo for various supersaturation ratios S.

S	2	3	4	5	6	7	8
r^* (Å)	17.0	10.7	8.5	7.3	6.6	6.0	5.7
g^*	695	174	87	55	40	31	26

terms of r^* directly. Noting from Table II that g^* is still as large as several hundred molecules even when we raise S beyond 2 (200% relative humidity if we return momentarily to the meteorological context), we see again how impossible it is to find critical embryos present unless S is quite high, the fundamental lesson learned from thermodynamic analysis of the nucleation problem.

Note that Eq. (4) is just Kelvin's equation for the enhanced saturation vapor pressure over a surface of small radius of curvature, now however appearing as the equation for the critical radius corresponding to a given supersaturated vapor pressure. These two seemingly different interpretations of (4) are not really distinct: In order that an embryo of radius r^* may be in metastable equilibrium with the surrounding vapor, it clearly must have just such a curvature as to raise its effective saturation vapor pressure from the value $p'(t)$ to the pressure p of the surrounding supersaturated vapor. Of the two interpretations, the critical-radius interpretation is much the more meaningful here, of course.

If we differentiate (2) a second time and examine the resulting sign, we find the second derivative to be intrinsically negative for $S>1$, confirming that our stationary value at r^* is, as depicted in Fig. 3, a *maximum*. Guggenheim gives what seems to this writer to be the most carefully and broadly formulated definition of a *metastable state*: any state which is stable with respect to *all* states differing only infinitesimally from the given state, but unstable compared with *some* other state differing finitely from the given state. The state associated with the maximum of the ΔF curve for any given value of S is thus, by Guggenheim's criterion, a metastable state. Curiously enough, we deal here with *a metastable state built, as it were, upon another metastable state*; for the supersaturated vapor itself exists in a metastable state before the beginning of formation of the critical embryo that is itself metastable with reference to the supersaturated vapor!

If we substitute the value of r^* from (4) back into the general equation (2) for ΔF, we evaluate the height of the free-energy activation barrier as

$$\Delta F^* = 16\pi\sigma^3/\{3(\rho RT\ln S)^2\} = 4\pi r^{*2}\sigma/3. \qquad (5)$$

In (5) ΔF^* is written in two equivalent forms, of which the latter, interpreted as one-third the product of the area of the critical embryo multiplied into the specific surface-free-energy can be carried over to nucleation of solids and used even in the event that the embryo must be assumed nonspherical for crystal-habit reasons. Gibbs used this latter form in his analyses, whence that form is sometimes identified as the Gibbs relation. Note that S enters (5) in such a way as to cause ΔF^* to fall off rapidly with rising S, at the same time that (4) shows r^* also diminishing with rising S. Equations (4) and (5) convey the fundamental thermodynamic moral of nucleation theory: *Raising the degree of supersaturation markedly enhances the probability that fluctuation processes will send some embryo over the top of the activation barrier in a given time because raising S reduces the embryo size r^* and the associated height ΔF^* of the top of the barrier.* Figure 3 displays this important point quite clearly. On the F-S plane of that model the projection of the skew curve passing through all the barrier summits has been plotted as the dashed curve (there labeled "ΔF_c", with c for "critical") that falls rapidly as the supersaturation ratio increases.

As will be elaborated in Part II of this discussion, a further enhancement of nucleation probability results from increasing S—namely the obvious effect of increased molecular bombardment rate attending any rise of vapor pressure. But this latter effect will be shown to be far less important than that cited in the preceding paragraph.

We may summarize all that has now been said: Sufficient increase of S (as we shall find, to values somewhere in the neighborhood of *five*) finally serves to raise the molecular bombardment rate and to reduce r^* and the attendant height of the activation energy barrier ΔF^* to such extent that the probability of some subcritical embryo fortuitously growing to supercritical size in a short time approaches unity. At that S value, homogeneous nucleation becomes an effective process, and phase transition can finally begin.

From the above statements it is clear that nucleation is *intrinsically a probabilistic event.* We may speak of the probable rate of appearance of critical embryos in a given volume of vapor at given S, or more meaningfully, we may speak of the average time we must wait for a single nucleation event in our given volume of vapor at supersaturation ratio S. The latter is the more physically realistic way to view the matter, inasmuch as it is generally true that once a *single* nucleation event has occurred *anywhere* within our system, it suffices to stabilize the *entire* system. In Part II we shall, however, find it mathematically more convenient to ask for the rate of appearance of nuclei under certain steady-state conditions contrived to simulate rather well the actual conditions preceding appearance of that first, crucial nucleus in our system.

An American Physicist's Year in Europe: Henry A. Rowland, 1875–1876

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Henry Augustus Rowland was born in Honesdale, Pennsylvania, in 1848, into a family of ministerial tradition, but he himself at an early age displayed an inclination toward and precocious gift for scientific experiment. The first twenty-seven years of his life were spent in self-preparation for a scientific career, although formally trained as a civil engineer at Rensselaer Polytechnic Institute, where he also taught physics. In 1875 occurred his "discovery" by Daniel C. Gilman, founder and first President of the new Johns Hopkins University, then in search of a suitable faculty. Now came his great opportunity, which Rowland eagerly seized upon, to spend a year in Europe, on salary from Johns Hopkins. Rowland's zealous and rewarding exploitation of this opportunity is the theme of this article. Rowland's letters and travel journal supply abundant material revealing how he traveled widely across Europe from Ireland to France, to Germany, Switzerland and Austria, and everywhere he visited the physical laboratories. He met many of the European men of science, inspected their facilities, and actually performed an important experiment in electromagnetism in Helmholtz's laboratory in Berlin. His critical commentary provides a unique survey of the state of physics in Europe at this time. In May, 1876, Rowland returned to America, unusually well-informed and self-assured, to assume the responsibilities of his new position as the first professor of physics in the Johns Hopkins University, where he built a distinguished scientific career for himself, broken off unhappily by his premature death in 1901.

THE essential interdependence of the international community of science is, of course, self-evident, despite and, indeed, because of its expanding role in an age of ideological controversy and national conflict. In an earlier time of less urgent necessity, natural isolation tended to restrict the intellectual and personal exchanges between Europe and America. The year spent in Europe by Henry A. Rowland in 1875–76 was then still an uncommon enough event to assume considerable significance of an ambivalent character. On the one hand, this experience was peculiarly timely and specifically invaluable in the development of one who was to become a major pioneer figure in American physics. It came at approximately mid-point in Rowland's life, when he was twenty-seven years old, and it completed, as it were, his education and transformation from an obscure and lonely young scientist of promise into a mature and self-confident practitioner of the relatively new science of physics at the just-established Johns Hopkins University.[1] The latter was itself a

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[1] For the earlier life of H. A. Rowland, see Samuel Rezneck, "The Education of an American Scientist: H. A. Rowland," in Am. J. Phys. **28**, 155 (1960); also "Com-