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Effect of the attractive potential of a drop in vapor phase nucleation

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The influence of the attractive potential between a drop and a vapor molecule on the rate of nucleation is investigated by considering its effect on the drop's capture cross section for a molecule. Previously unsuspected effects that may occur under certain conditions are revealed. In particular, as the carrier gas pressure is increased, there may be a strong decrease in the nucleation rate. [S1063-651X(96)02410-5]

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I. INTRODUCTION

The theory of vapor phase homogeneous nucleation has evolved since the 1930's [1–3] and has been continually revised, but no change has been made in the manner of estimating the average rate at which molecules collide with a drop. It has been customary to calculate this rate by using the surface area of the drop. In the present paper we examine and revise this aspect and uncover the consequences.

In retrospect, the physical basis of our results is easy to understand, and it is worthwhile to present it at the outset. A drop of the condensed phase must accumulate a critical number of molecules, to become unstable with respect to growth. Each absorption of a molecule is enhanced by the drop's attractive potential. At low supersaturations the critical drop size is large, and so the enhancement is applied many times during the drop's growth to this size. This makes the nucleation rate sensitive to the enhancement. The enhancement, in turn, is sensitive to the density of the vapor and carrier gas, because the effect of the drop's attractive potential is reduced by encounters of incoming molecules with other vapor or carrier gas molecules, an effect that may be referred to conveniently but inaccurately as a ''screening effect.'' This sensitivity may result in a dependence of the nucleation rate on the carrier gas pressure.

In Sec. II (and Appendix A) we revise the conventional expression for the impact rate of pointlike molecules on a spherical drop. In Sec. III (and Appendix B) we discuss possible consequences for steady state nucleation.

II. MOLECULE-DROP CROSS SECTION

We begin with the conventional expression for the average rate β at which vapor molecules, having a Maxwell distribution, hit a drop

$$\beta=\beta^{(s)}\equiv\left(\frac{kT}{2\pi m}\right)^{1/2}\rho\langle s\rangle, \tag{1}$$

where m is the molecular mass, ρ is the number density of vapor molecules, k the Boltzmann constant, T the temperature, and the superscript s denotes an estimate based on the average surface area of the drop. This area is estimated as $\langle s\rangle=4\pi R_d^2$; $R_d=(3j/4\pi\rho_{\mathrm{con}})^{1/3}$, where R_d is the radius of the drop, ρ_{con} is the molecular number density in the condensed phase, and j is the number of molecules in the drop. We will revise Eq. (1) taking into account the fact that a drop of interest for nucleation is typically small, even in comparison to the mean intermolecular distance in the vapor.

First, consider a drop devoid of its attractive potential, i.e., such that the potentials of all its molecules are arbitrarily cutoff at the drop surface. Set the sticking coefficient to unity, as usual. The rate at which molecules accumulate in this drop is then

$$\beta^{(c)}=\rho\langle v\sigma\rangle=\rho\langle v\rangle\langle\sigma\rangle=\left(\frac{8kT}{\pi m}\right)^{1/2}\rho\langle\sigma\rangle, \tag{2}$$

where the superscript c denotes ''cutoff'' and where $\langle\sigma\rangle$ is the drop cross section σ averaged over all directions, and $\langle v\rangle$ is the average molecular velocity. For a spherical drop of radius R_d, $\langle\sigma\rangle=\pi R_d^2=\langle s\rangle/4$, and $\beta^{(c)}(j)=\beta^{(s)}(j)$. For other (nonspherical) shapes, $\langle\sigma\rangle\neq\langle s\rangle/4$. 4 $\langle\sigma\rangle$ and $\langle s\rangle$ have quite different geometric characters. For example, $\langle\sigma\rangle$ is much less sensitive to shape fluctuations. To realize this, it is enough to imagine a bump on the surface of the sphere; the bump is not even visible from most directions.

Next consider the more realistic situation without an artificial cutoff. The closest a molecule with speed v, when it is infinitely far from the center of a central field $U(r)$, comes to the center of that field is given by $r=r_c$, where r_c is a largest root of the following equation [4]:

$$U(r_c)+\frac{M^2}{2mr_c^2}=\frac{mv^2}{2}, \tag{3}$$

where $M=mRv$ is the angular momentum of the molecule while R is the ''impact parameter'' for the encounter of the molecule with the field which we take to be that of the drop. The molecules with $R>R_{\max_1}(v)$ with certainty will not touch the drop, where $R_{\max_1}(v)$ is given by the value of R that satisfies Eq. (3) with R_d substituted for r_c. We find $R^2_{\max_1}(v)=R_d^2(1-[2U(R_d)/mv^2])$. If $R<R_{\max_1}(v)$ was the only restriction, the impact rate β would be equal to

$$\pi\rho\langle vR^2_{\max_1}(v)\rangle=\left(1-\frac{U(R_d)}{kT}\right)\beta^{(c)}, \tag{4}$$

where the averaging is according to the Maxwell distribution. Equation (4) does not depend on the form of the potential; only the magnitude of $U(R_d)$ is involved. However, Eq.

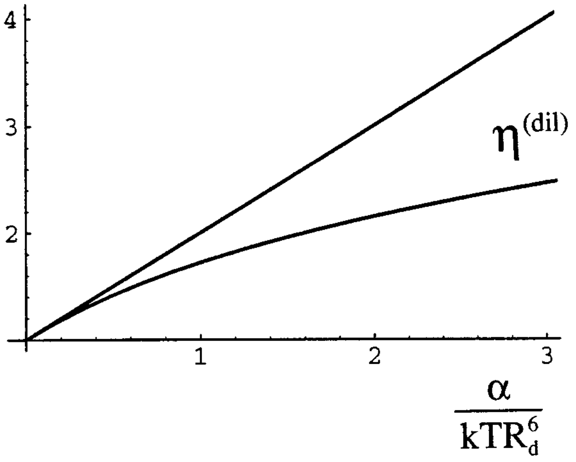

FIG. 1. The enhancement factor η vs (α/kTR_d^6) [for the potential $U(r)=-\alpha/r^6$] and its tangent $(1+[\alpha/kTR_d^6])$.

(3), for a pair of variables v,R where $R=R_{\max_1}(v)$, may have another root, larger than R_d, for small enough v. For a potential $U(r)=-\alpha/r^n$, $n>2$, the resulting restriction is $R<R_{\max}(v)$, where $R_{\max}(v)=R_{\max_1}(v)$ at $v>v_0$ and $R_{\max}(v)=R_{\max_2}(v)=[n(n/n-2)^{(n-2/2)}(\alpha/mv^2)]^{1/n}$ at $v<v_0$, and $v_0\equiv[(n-2)/(\alpha/mR_d^n)]^{1/2}$. (For more details see Appendix A.) The resulting impact rate, for this case, is

$$\beta^{(\mathrm{dil})}=\pi\rho\langle vR_{\max}^2(v)\rangle=\left[u^{2/n}\gamma\left(1-\frac{2}{n},u\right)+e^{-u}\right]\beta^{(c)}, \tag{5}$$

where $u\equiv(n-2/2)(\alpha/R_d^n kT)$ and γ is the incomplete gamma function [5]. The label (dil) indicates that the result is limited to a vapor, sufficiently dilute so that intermolecular collisions do not have to be considered along the molecular trajectory. The small u expansion

$$\frac{\beta^{(\mathrm{dil})}}{\beta^{(c)}}=1+\frac{2}{n-2}u+O(u^2)$$

has the form of the right-hand side of Eq. (4). For $n=6$ our enhancement factor

$$\eta^{(\mathrm{dil})}\equiv\frac{\beta^{(\mathrm{dil})}}{\beta^{(c)}}$$

is shown in Fig. 1.

$|U(R_d)|$ may be estimated as the depth of the drop's potential well, which, in turn, is nominally of the order of the depth of the intermolecular pair potential. Depending on the substance, this is typically of order 10–1000 K (in units of temperature) [6]. Thus Eq. (5) indicates that $\beta^{(\mathrm{dil})}$ may be of order of $\beta^{(c)}$ or several fold larger. (For a review of typical experimental conditions, see [7].)

In case the reader feels uncomfortable about such an enhancement of the conventional $\beta^{(s)}$, we point out that, the larger the drop, the more dilute must the surrounding gas be for the enhancement to be valid. Otherwise, as we discuss in Sec. III, the conventional result is recovered. And if one imagines, for the sake of understanding, a macroscopic sphere (''drop'') in a vacuum, then the dispersion potential $\sim-(1/r^6)$ must be corrected at large distances [6]. Also, small angle deflections of slow particles in the potential may require quantum corrections.

III. STEADY STATE NUCLEATION AND A CARRIER GAS EFFECT

A refined estimate of β can now be used in the standard expression [2,8–10] for J the steady state rate of nucleation, given by

$$J=\rho\left(\sum_{j=1}^{\infty}\frac{1}{\beta(j)f(j)}\right)^{-1}, \tag{6}$$

in which $f(j)=\Pi_{l=1}^{j-1}[\beta(l)/\gamma(l+1)]$ and $f(1)\equiv1$. Here $\beta(l)$ is β for a drop of l molecules, while $\gamma(l)$ is the average rate at which molecules evaporate from the drop. In principle, γ may be directly calculated from a drop model and substituted into Eq. (6). Such an approach was attempted recently by Nowakowski and Ruckenstein [11].

It is well known that the dominant terms in Eq. (6) correspond to ''critical'' drops having values of j typically in the range 10 to 100 [1,2]. As a result the factor $\eta^{(\mathrm{dil})}$, which enters $f(j)$ through the product of β's, appears in J of Eq. (6) raised to a high power.

This effect is nearly eliminated in the conventional theory, in which γ's are obtained through an application of the principle of detailed balance that involves evaluating the equilibrium distribution of drops (see, e.g., [2,8,9]). Then, f is expressed as

$$f(j)=\left(\prod_{l=1}^{j-1}\frac{\beta(l)}{\beta_e(l)}\frac{\gamma_e(l+1)}{\gamma(l+1)}\right)\exp\left(-\frac{G(j)}{kT}\right), \tag{7}$$

where $\exp\{-[G(j)/kT]\}=\Pi_{l=1}^{j-1}[\beta_e(l)/\gamma_e(l+1)]$, and the subscript e (equilibrium) refers to quantities corresponding to the saturated vapor. $G(j)$ is the Gibbs free energy that must be supplied (i.e., the reversible work that must be performed) to excise a j drop from liquid coexisting with vapor. In the classical theory [3] this free energy is estimated using the ''capillarity approximation'' such that $G(j)=\varrho s(j)$ where ϱ is the bulk surface tension. (This capillarity approximation can be substantially improved [9,12] or replaced by a density-functional method [13] or by a fully molecular approach [14].) Now, since $[\beta^{(\mathrm{dil})}(l)/\beta_e^{(\mathrm{dil})}(l)]=(\rho/\rho_e)$ and $\gamma(l)=\gamma_e(l)$, based on the excellent approximation that γ depends only on the internal properties of the drop [2,11,15] the factor $\eta^{(\mathrm{dil})}$ does not appear in Eq. (7). This factor therefore appears only once in each term of Eq. (6).

However, another situation arises when the vapor (or the vapor-carrier gas mixture [7]) is dense enough, or the critical drop size is large enough. We now discuss the possible consequences of a crossover of β from $\beta^{(\mathrm{dil})}$ to $\beta^{(c)}$ due to the influence of the gas surrounding the drop: $\beta=\eta\beta^{(c)}$, where $1\leq\eta\leq\eta^{(\mathrm{dil})}$. Unlike $\eta^{(\mathrm{dil})}$, η generally depends on the number densities ρ and ρ_1 of the vapor and carrier gas, respectively. Apparently, $\eta=\eta(l;\rho;\rho_1)$ decreases with an increase of ρ or ρ_1 (we also indicate that η depends on the droplet size l). Now

$$\frac{\beta(l)}{\beta_e(l)}=\frac{\rho}{\rho_e}\,\frac{\eta(l;\rho;\rho_1)}{\eta(l;\rho_e;\rho_1)}<\frac{\rho}{\rho_e},$$

and $f(j)$ in Eq. (7) must be corrected by the multiplier

$$\prod_{l=1}^{j}\frac{\eta(l;\rho,\rho_1)}{\eta(l;\rho_e,\rho_1)}<1.$$

Thus the nucleation rate is somewhat below that predicted by the classical theory. However, typically the carrier gas density is much higher than the vapor density [7].

In the remainder of this paper we discuss the dependence of η on ρ_1, that may result in an effect that should be observable directly by experiment and be qualitatively independent of any particular theory, namely, the effect of carrier gas on the nucleation rate. With $\beta(l;\rho,\rho_1)=\eta(l;\rho_1)\beta^{(c)}(l;\rho)$ and γ independent of ρ_1, Eqs. (6) and (7) indicate that the steady state nucleation rate $J=J(\rho,\rho_1)$ depends on ρ_1 in the following manner:

$$J(\rho,\rho_1'')\sim\left(\prod_{l=1}^{j_{\rm cr}}\frac{\eta(l;\rho_1'')}{\eta(l;\rho_1')}\right)J(\rho,\rho_1'), \qquad (8)$$

where $j_{\rm cr}$ is the critical drop size. In arriving at Eq. (8) we took γ to be independent of ρ_1 because the gas densities are assumed to be high enough such that thermal equilibration by the carrier gas is established [16]. Equation (8) predicts a unidirectional effect, i.e., a decrease in nucleation rate (and thus an increase in critical supersaturation) with an increase of carrier gas density.

The full correction $\langle v[R_{\max}^2(v)-R_d^2]\rangle$ in Eq. (5) was derived only because the drop was allowed to capture all those molecules moving slowly enough, independent of impact parameter. But for a typical rapidly damped intermolecular potential, e.g., $u(r)\sim -r^{-6}$, the trajectory of a molecule A towards the drop is interrupted (to a first approximation, randomized) as soon as another molecule B is closer to A than the drop.

Thus, qualitatively, $\eta\approx 1$ if the average carrier gas intermolecular distance $\sim\rho_1^{-(1/3)}$ is much smaller than the critical droplet size $R_{\rm cr}$, and $\eta\approx\eta^{(\rm dil)}$ if $\rho_1^{-(1/3)}\ll R_{\rm cr}$. (We provide an estimate of the deviation of η from $\eta^{(\rm dil)}$ in Appendix B.) The important ratios are [see Eq. (8)]

$$\frac{\eta(l;\rho_1'')}{\eta(l;\rho_1')}\approx 1$$

if $\rho_1',\rho_1''\ll R_{\rm cr}^{-3}$ or $\rho_1',\rho_1''\gg R_{\rm cr}^{-3}$. Qualitatively from Eq. (8) [see also Eq. (B3)], we expect an onset of the strong carrier gas effect under conditions such that the average carrier gas intermolecular distance is comparable to the critical droplet size. The classical estimate for the latter is $R_{\rm cr}\approx(2\varrho/\rho_{\rm con}kTLnS)$, where $S=(\rho/\rho_e)$ is the degree of supersaturation of the vapor.

The effect (a decrease in the nucleation rate with an increase in carrier gas pressure) might be observed in experiments with vapors with molecules of large enough size, if nucleation is observable at low enough supersaturations.

In fact, there is evidence of such an effect [17–19]. According to [17], the critical supersaturation of 1-pentanol, 1-butanol, 1-propanol, and even of ordinary and heavy water increases with increasing carrier gas (helium) concentration. The effect increases with decreasing temperature or with increasing molecular weight of the alcohol. The same trends were observed recently for the methanol, ethanol, 1- and 2-propanols, with helium and hydrogen as carrier gases [18]. In [19] the effect has been observed for the nucleation of n-nonane in helium and argon [19]. Remarkably, it has not been seen at low carrier gas (argon) pressures (less than 1 bar, in the nucleation of n-butanol, methanol, and water [20]). It is also interesting that there is evidence of carrier gas influence in the nucleation of water vapor at low supersaturations [17,21], but the effect has not been found at higher supersaturations ($S>7$) [22]. Also, for water the effect has not been found at low carrier gas (nitrogen) pressures [23].

These experimental observations cannot be explained by a weak influence of the carrier gas and vapor nonideality on the equilibrium distribution of drops [19,25], and it is possible that they may be related to the ''screening'' effect discussed here, which has an entirely different (and kinetic) origin.

To check this possibility, we estimated the carrier gas pressure

$$P=\frac{kT}{\dfrac{4\pi}{3}R_{\rm cr}^3}$$

which should produce this effect. To estimate

$$R_{\rm cr}\approx\frac{2\varrho}{\rho_{\rm con}kTLnS},$$

we used the critical supersaturations for methanol, ethanol, n-propanol, and i-propanol from [18,17,26,27] and the surface tensions ϱ of these substances from [28,29]. Making polynomial interpolations of these values in temperature, we find, for instance, that at $T=350$ K, the P's for all these substances lie within the range 1–2 bar (whereas the effect was investigated in [18] at total pressures extending from several bar to 40 bar). At $T=330$ K all P lie within 2–3 bar. And at $T=380$ K all P lie within 0.3–0.8 bar. For these large temperatures the effect is, actually, ''saturated'': a change in the carrier gas density from ρ_1' to ρ_1'' should not much change the ratios $[\eta(l;\rho_1'')/\eta(l;\rho_1')]\approx 1$ when both $\rho_1',\rho_1''\gg\rho_{\rm con}/l$. This may explain why the effect is found to be less pronounced at higher temperatures [18].

Concluding this section, we must mention that a slight influence of carrier gas due to nonisothermal effects has been predicted in [16]. At low carrier gas pressures the latent heat of condensation cannot be easily removed from a newly formed drop. This effect should have a weak opposite trend (an increase of the nucleation rate with increasing carrier gas pressure), must show itself at very low carrier gas pressures, and has probably been observed [24].

IV. CONCLUSION

In conclusion, we have argued that the cross section $\beta^{(\rm dil)}$ of a small drop in a dilute vapor-gas mixture may be several times larger than the conventional $\beta^{(s)}$ Eq. (1). We have indicated and discussed the adjustments in nucleation theory required by this change in cross section. Furthermore, we have discussed the sensitivity of β to the density of the gas-vapor mixture. Screening affects the impact of vapor molecules on the drop approximately $j_{\rm cr}$ times, during growth to the critical size, Eq. (8). Among other things it may appear as an effect of a carrier gas on the rate of nucleation (decrease of the rate with increasing the carrier gas pressure).

We have not discussed the question of sticking probabilities for very small drops. A sticking probability is larger for a slower molecule and the average energy of a molecule colliding with a spherical drop is $[\langle v\sigma(mv^2/2)\rangle/\langle v\sigma\rangle]$. The ''additional'' molecules considered in this paper, i.e., the molecules that would not collide with the droplet in the absence of an attractive potential, have a lower average energy. Thus sticking probabilities that are not unity can only enhance the effect discussed in this paper.

We also have not considered the internal degrees of freedom of the drop or the molecules. These may absorb some angular momentum and so increase $\beta^{\rm (dil)}$. The revision may be important in the case of vapors of molecules of larger size. These are of special interest in view of Eqs. (8) and (B3).

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APPENDIX A

The radial velocity v_r of a particle in a spherical field $U(r)$ is given by [4]

$$v_r^2(r)=v^2\left(1-\frac{R^2}{r^2}\right)-\frac{2}{m}\,U(r). \tag{A1}$$

A particle reaches a drop of radius R_d if it has initial velocity v and impact parameter R such that $v_r^2(r)>0$ for all $r\geqslant R_d$. [Equation $v_r^2(r)=0$ gives Eq. (3).] Thus

$$v_r^2(R_d)>0 \tag{A2}$$

is the necessary condition. [It is equivalent to $R<R_{\max_1}(v)$, see Sec. II.] We define R_d as the distance at which the attractive tail of $U(r)$ is terminated by a typical sharp repulsive core. So, all particles (v,R) which satisfy Eq. (A2) and

$$[\partial_r v_r^2(r)]_{r=R_d}\geqslant 0 \tag{A3}$$

will certainly reach the drop. For a potential $U(r)=-(\alpha/r^n)$ Eq. (A3) reads $v\geqslant v_1(R)$, where $v_1^2(R)=(n\alpha/mR_d^{n-2}R^2)$. If $[\partial_r v_r^2(r)]_{r=R_d}<0$, we must add the restriction

$$\min\, v_r^2(r)>0. \tag{A4}$$

For the potential $U(r)=-(\alpha/r^n)$ Eq. (A4) reads $v<v_2(R)$, where $v_2^2(R)=n(n/n-2)^{(n-2/2)}(\alpha/mR^n)$. [The restriction is equivalent to $R<R_{\max_2}(v)$, see Sec. I.] Thus the restriction $v<v_2(R)$ must be taken into account at $v<v_0$, where v_0 is defined as $v_0=v_2(R_0)$; $v_1(R_0)=v_2(R_0)$. It is straightforward to find $R_0=\sqrt{(n/n-2)}R_d$, v_0 is as in Sec. I, and the restriction $v<v_2(R)$ is stronger than that in Eq. (A2) at $v<v_0$.

APPENDIX B

The full correction $\langle v(R_{\max}^2(v)-R_d^2)\rangle$ in Eq. (5) was derived only because the drop was allowed to capture all those molecules moving slowly enough, independent of impact parameter. But for a typical rapidly damped intermolecular potential, e.g., $u(r)\sim -r^{-6}$, the trajectory of a molecule A towards the drop is interrupted (to a first approximation, randomized) as soon as another molecule B is closer to A than the drop. Thus, our cross section must be restricted to molecules with an impact parameter smaller than some cutoff, $R_{\rm cut}\sim\rho_1^{-(1/3)}$ (we consider the number density of the carrier gas molecules to be much larger than that of the vapor molecules). We can estimate $R_{\rm cut}$ as following. In time Δt a molecule A encounters approximately $\rho_1\langle v_g\sigma_g\rangle_g\Delta t$ molecules B (carrier gas molecules), where σ_g is the corresponding cross section, and the average $\langle\cdots\rangle_g$ is taken over the Maxwell distribution of the velocities v_g of the molecules B. Thus $R_{\rm cut}$ is determined by

$$1\sim\rho_1\langle v_g\sigma_g\rangle_g\Delta t \tag{B1}$$

with $\Delta t\sim(R_{\rm cut}/v)$. $\sigma_g=\pi R_g^2$ is the cross section for a deflection of the trajectory of the molecule A stronger than that caused by the drop. Since a small acquired momentum due to the potential α/r^n is proportional to α, and inversely proportional to the relative velocity and to the nth power of the impact parameter [4], we can estimate R_g by equating $(\alpha_g/v_gR_g^6)\sim(\alpha/vR_{\rm cut}^6)$. For simplicity, (α_g/r^6) represents the potential between B and A and (α/r^6), the potential between the drop and A (although when A is very close to the drop, the potential cannot have this simple form). We approximate the velocity of B relative to A by v_g (A is slow) and the velocity of A relative to the drop by v (the drop is slow). Substitution of $\sigma_g\sim(\alpha_g v/\alpha v_g)^{1/3}\pi R_{\rm cut}^2$, into Eq. (B1) leads to $R_{\rm cut}=R_0(v^{2/9}/\langle v\rangle^{2/9})$, where

$$R_0\sim\left(2^{1/3}\pi^{5/6}\Gamma\left(\frac{11}{6}\right)\left(\frac{\alpha_g m}{\alpha m_g}\right)^{1/3}\rho_1\right)^{-1/3}. \tag{B2}$$

Here m_g is a mass of the carrier gas molecule. This dependence of $R_{\rm cut}$ on v is reasonable since a slower molecule A has a larger probability of experiencing an encounter with carrier gas molecules B on its way to the drop. It is easy to see, however, that the estimate, with the small exponent 2/9 and with the exponents 1/3 in Eq. (B2) is qualitatively the same as the guess $R_{\rm cut}\sim\rho_1^{-(1/3)}$.

To estimate the order of the effect of the carrier gas on our enhancement factor η, we impose an additional restriction $R<R_{\rm cut}(v)$ [compare with $R_{\max_1}(v)$ and $R_{\max_2}(v)$]. Notice that the velocity $v_{\rm cut}$ defined by $R_{\rm cut}(v_{\rm cut})=R_{\max_2}(v_{\rm cut})$ is

related to v_0 (see Sec. II) as $(v_{\mathrm{cut}}/v_0)^2 = (3^9/2^7\pi^2)^{1/5}(kTR_d^6/\alpha)^{2/5}(R_d/R_0)^{18/5}$. Here the key ratio is R_d/R_0, which is essentially the ratio of the drop size to the average intermolecular distance in the carrier gas. According to the classical estimate for the critical drop radius $R_{\mathrm{cr}} \approx 2\varrho/\rho_{\mathrm{con}}kTLnS$, where ϱ is the bulk surface tension of the condensed phase, R_{cr} may be of order 10–15 Å or more, for a low enough ρ_{con} (i.e., for large enough vapor molecules) [1]. While

$$\rho_1^{-1/3} \approx \left(\frac{T}{300\ \mathrm{K}} \Big/ \frac{P_1}{1\ \mathrm{Bar}}\right)^{1/3} 35\ \text{Å},$$

where P_1 is the carrier gas pressure. Thus at moderate carrier gas pressures $v_{\mathrm{cut}} < v_0$ and we obtain [analogously to Eq. (5), but taking into account $R < R_{\mathrm{cut}}(v)$]

$$\eta = \eta^{(\mathrm{dil})} - \frac{3^5}{2^{4/3}\pi^{2/3}}\left(\frac{\alpha}{kTR_d^6}\right)^{4/3}\left(\frac{R_d}{R_0}\right)^6 \qquad \text{(B3)}$$

to the first order in $(mv_{\mathrm{cut}}^2/2kT) = (3^9/4\pi^2)^{1/5}(\alpha/kTR_d^6)^{3/5}(R_d/R_0)^{18/5} < 1$.

Unlike $\eta^{(\mathrm{dil})}$ (which is at least universal at small u), the ‘‘screened’’ η depends strongly on the functional form of the potential between the drop and the vapor molecule, and also, to a lesser extent, on the type of potential between the carrier gas molecule and the vapor molecule. These potentials are not generally known, except for the noble gases [6]. Thus we cannot make quantitative estimates based on Eq. (8). However, qualitatively from Eqs. (8) and (B3), we expect an onset of the strong carrier gas effect under conditions such that the average carrier gas intermolecular distance $\rho_1^{-(1/3)}$ is comparable to the critical droplet size $R_{\mathrm{cr}} \approx 2\varrho/\rho_{\mathrm{con}}kTLnS$.

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