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Can the spinodal decompositon generate a resistivity anomaly in a binary alloy?

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Abstract

In this work we discuss the real dependance that the residual electric resistivity has on the aging time, when the spinodal mechanism is responsible for the decomposition of *a* binary doy. We use the isotropic aproximation for the resistivity and linear, as well as nonlinear theory of spinodal decomposition, Our results, in both cases, show an increment in the resistivity up to a point which is determined, in the nonlinear theory, by the critical amplitude of fluctuation of concentration, which disagees with other results reported in the literature. With these results we show that this mechanism by itself cannot explain the anomaly exhibited by electrical resistivity.

Key words: Binary *doy*; residual electric resistivity; spinodal decomposition.

¿Puede la descomposición spinodal generar una anomalía en la resistividad de una aleación binaria?

Resumen

En este trabajo discutimos la dependencia real que la resistividad eléctrica residual tiene con el tiempo, cuando el mecanismo spinodal es el responsable de la descomposición de una aleación binaria. Hemos usado la aproximación isotrópica para la resistividad *y* tanto la teoría de descomposición spinodal lineal como la no lineal. Nuestros resultados en ambos casos, muestran un incremento en la resistividad en el punto critica, el cual se determina, en la teoría no lineal. por la amplitud crítica de las fluctuaciones de concentración. lo cual está en desacuerdo con otros resultados reportados en la literatura. Con estos resultados demostraremos que este mecanismo no puede por si mismo explicar la anomalía exhibida por la resistividad eléctrica,

Palabras claves: Aleación binaria; descomposición spinodal; resistividad eléctrica residual.

Introduction

The answer to the question raised by this paper must be sougt in the theory of Gibbs fluids (1), where the fundamentals of the evolution of a single phase system towards a multiphases system are well explained. Relying on this theory and depend-

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ing upon the stability of the system under study, we can diferenciate two mechanisms of decomposition: the germination and growth mechanism and the spinodal decomposition mechanis, (SD). These mechanisms evolve from metastable and unstable solid solutions following down-hill and up-hill diffusion processes, respectively.

The SD theory has been applied to solid solutions by many authors, among the first being Cahn (2,3) and Hillert (4), who predicted a decomposition of solid solution with the preferential growth of the concentration fluctuations around a so-called maximun wavelength. Cahn (5), Cook *et al.* (6,7), Langer *et al.* (8), Binder *et al.*(9) and Tsakalakos (10) have made complete and sophisticated presentations of this theory.

The Spinodal decomposition starts from a solid solution in the unstable regime, which during a brief time has a structure with interconnectivities characteristic of this type of decomposition (11). This structure disappears after some time, to allow the formation of precipitates. The decomposition sequence is an ideal process for examination by electric resistivity studies. As a matter of fact, several experimental works have been done of binary alloy of aluminium, with impurity concentrations and aging temperatures which suggest that they are in the SD regime. In those examples, the temporal evolution of the resistivity shows a so-called abnormal behaviour, because it increases, reaches a maximun and then decreases to a plateau (12 -14).

In general, few authors have related this behaviour to the mechanisms suggested in Gibbs theory. Furthermore, the way of evaluating the participation of the dispersive processes supposes a decomposition by germination and growth, where the conduction electrons are scattered by Guinier-Preston zones which evolve until they coalesce (15 - 18), are among the few author who have said that the abnormal behaviour is characteristic of the SD, and have reinforced their ideas with studies using different techniques. But their conclusions do not seem to be unique to this mechanisms and, on the contrary, Radomsky and Löffler (19) have presented a paper concerning the resistivity in this regime and predicting a monotonic decrease of the residual resistivity, in complete disagreement with the ideas of the Poitiers group. Earlier, Kolomets *et al.* (20) had come to the conclusion that the residual electric resistivity of a nonordered binary alloy decreases when a periodic modulated structure appears, and that such a decrease is proportional to the square of the amplitude of the concentration fluctuation.

In this work we use Boltzmann's theory for the treatment of the electrical resistivity. For the DS study we use Cahn's (2) y Tsakalakos's (10) non-linear theory. We have chosen the binary system Al - Zn for the application of our study since its wide gap miscibility makes it an ideal system.

Method

The methods to study the real behaviour of the resistivity during the process of spinodal decomposition should combine transport theory and thermodynamic theory. The former should be used to define an adecuate relation for the electrical resistivity and the latter should introduce the decomposition process. Binder *et al.* (9), in his general treatment of phase transitions in binary systems, as well as Mimault *et al.* (18), follow this scheme, but their work show details which do not agree with the spinodal decomposition process. In the following section we give a brief summary of the spinodal decomposition theory and resistivity theory.

Electric Resistivity

Boltzmann's theory, based on the free electron model, using the first Born approximation and supposing isotropy, states that the time evolution of the electrical resistivity is given by

$$\rho(t) = c' \int \langle W^2(q) S^2(q,t) \rangle \, q^3 dq \qquad [1]$$

where c' is a constant, W^2S^2 is the elastic transition probabilility between the electronic states $\underline{k}$ and $\underline{k}'$ which are related with wavevector $\underline{q}$ by $\underline{q} = \underline{k} - \underline{k}'$, W is named the form factor and S is the structure factor.

The form factor, W(q), will be considered in the point ion model as was done in previous work (16,21). The structure factor, S(q,t), is the Fourier transform of the atomic correlation function, and it is through this factor that relation [1] incorporated in DS mechanism.

Usually, the maximum number of integrations in [1], is on Fermi's surface, but that is not the case in thips work since the process of spinodal decomposition ocurrs for a characteristic range of wavelengths bounded in the upper limit by the vector q_c, which we will define later in this work. The choice of lower limit equal to inverse of the mean free path of electrons, λ^{-1} (9), might have some influence in the magnitude of the resistivity provide that q_c be of the same order of magnitude as λ^{-1}.

The parameter which finally is of our interest is $\delta\rho(t)$, which is the relative change, with respect to initial resistivity $\rho(0)$, of the resistivity during the process of spinodal decomposition. The calculation is done inmediately after a hypothetic quenching from a monophase to a polyphase region

$$\delta\rho(t) = \frac{\int \langle u^2(q) s^2(q,t) \rangle \, q^3 dq - \int \langle u^2(q) s^2(q,0) \rangle \, q^3 dq}{\int \langle u^2(q) s^2(q,0) \rangle \, q^3 dq} \qquad [2]$$

Spinodal decomposition

In this work we calculate the structure factor in both theories: the linear theory of Cahn (2) and the non-linear theory of Tsakalakos (10). In both theories its necesary to know beforehand the Gibb's free energy in order to study the kinetics of the decomposition. This is done through the evolution of the flux of impurity atoms, J, which is due to a change in the local concentration. The time evolution of the concentration C, obeys the relationship (2,7):

$$\frac{\delta c}{\delta t} = -\nabla . J = V_0 M \left\{ \left[f' + \frac{2\eta^2 - E}{1-\nu} \right] \nabla^2 c - 2\kappa \nabla^4 c - \nabla \left[\frac{\delta^2 \kappa}{\delta c^2} (\nabla c)^3 + 4 \frac{\delta \kappa}{\delta c} \nabla c \nabla^2 c \right] \right\} \qquad [3]$$

where M is the atomic mobility, f" is the second derivative of the free energy evaluated at the average concentration, C_o, V_o is the atomic volume, κ is the gradient-energy coefficient, η is d(ln a)/dC, a is the lattice parameter, E is Young's modulus and ν is the Poisson coefficient.

Among the infinite solutions of the diffusion equation, [2], the one with greater acceptance is:

$$C(r,t) - C_0 = \int A(q,t) \cos(q.r) \, d^3q \qquad [4]$$

The amplitude of the fluctuation of concentration A(q,t) is proportional to the structure factor S(q,t) and obeys a differential equation similar to [3].

$$S^2(q,t) \; \alpha \; A(q,t) \, A^*(q,t) \qquad [5]$$

In the generalized Cahn's theory, equation [2] simplifies if we take both the energetic gradient κ and the elastic coefficients ν to be independent of concentration. This restricts solution [3] to early stage the decomposition process, with amplitude defined by

$$A(q,t) = A(q,0) \exp[R(q)t] \qquad [6]$$

In the non-linear theory we choose the approximate solution of Ditchek (22), where A(q,t) is given by

$$A(q,t) = A_c(q)\tanh\{[A(q,0)/A_c(q)]\exp[R(q)t]\} \quad [7]$$

In these equations, A(q,0) represent the amplitude of the fluctuations at t=0, A_c is the critical amplitude defined as:

$$A_c(q) = [1-(q/q_c)^4]^{\sqrt{1/2}} \quad [8]$$

where q_c is the q value which, being non-zero, cancels out the amplification factor R(q).

Only after Aq,t) grows will the thermodynamic conditions be adequate to favor a spinodal decomposition. This requires that the amplification factor R(q) be greater than zero.

$$R(q)=-MV_o q^2 [f'' + 2q^2\kappa + 2\eta 2E/(1-\nu)] \quad [9]$$

The initial amplitude A(q,0) depends fundamentally upon the quenching temperature and the speed of the quenching process. A(q,0) can be taken as independent of q, however, a better result is obtained by taking the gaussian distribution of Ditchek *et al.* (23).

$$A(q,0)=a\exp\{0.5[(q-\sqrt{2}\,q_c)/w]^2\} \quad [10]$$

a and w being parameters of the gaussian distribution. In this way relations [5] to [10] define completely the structure factor in the DS theory.

Results

The time evolution of the residual resistivity given by [2] has been numerically evaluated for different alloy Al-Zn alloys. Here we only report the Al - 18%at.Zn alloy, since the results obtained with this alloy are representative of all the other alloys studied (22%, 35%). The thermodynamic parameters which appear in relation [9] depend upon the concentration of solute and upon temperature. These parameters are generally taken from phenomenological work (24). In our case we use for f" and $2\eta^2E/(1-\nu)$ the same expresion of Simmich *et al.* (25).

Table 1

Parameters used in the evaluation of δρ(t)

C_o	18%	
T	323 K	
D_0^{Al}	$1.71\text{x}10^{-4}$ m^2/s	(Bokstein. 1978)
Q^{Al}	34.0 Kcal/mol	(Bokstein. 1978)
D_0^{Zn}	0.259x10-4 m2/s	(Erdelyi *et al.1978*)
Q^{Zn}	28.86 Kcal/mol	(Erdelyi *et al.*1978)
M	$1.97174\text{x}10^{-5}$ Kg^{-1}s	
f"	$1.092\text{x}10^{10}$ N m^{-2}	
$\frac{2\eta^2E}{1-\nu}$	$1.361\text{x}10^{8}$ N m^{-2}	
κ	1.00x(10^{-9} or 10^{-11}) N	

The atomic mobility will be evaluated following Houston *et al.* (26).

$$M= C_0(1-C_0)\{[C_0 D_0^{Al}\exp(-Q^{Al}/KT) + (1-C_0)D_0^{Zn}\exp(-Q^{Zn}/KT)]/KT\} \quad [11]$$

where D_0^{Al} and D_0^{Zn} represents the pre-exponential accompaning Arrhenius's expresion of the diffusion coefficient. Q^{Al} and Q^{Zn} are the activation energy for diffusion.

In the Table 1, we show the data used in our calculations. The κ values are generally ambiguous in the literature so we left it as a free parameter which could vary around the value suggested by Rudman *et al.* (24) for Al-22%at Zn alloy. The choice of different κ values produces a modification in q_c, being greater the smaller we choose κ. This effect will favor large angle scattering.

In Figure 1 we plot, for different κ values, the amplification factor R(q) in terms of the variable X ($X=q/K_F$; K_F is Fermi's wave vector) for the Al-18%at.Zn alloy. In the right hand side scale we choose $\kappa=10^{-9}$ N,

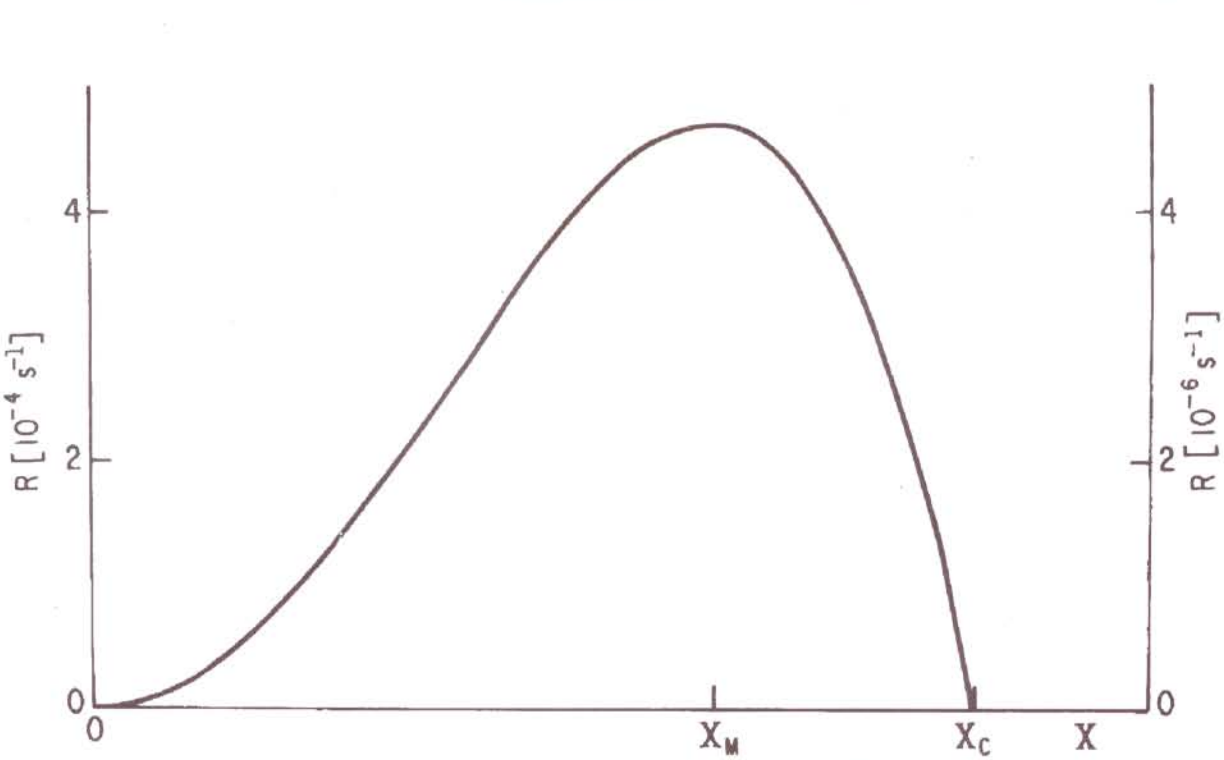

Figure 1. Amplification factor R versus $X=q/K_F$. Right scale for $\kappa=10^{-9}$N, $X_M=0.0938$, $X_C=0.1327$. Left scale for $\kappa=10^{-11}$, $X_M=0.938$. $X_C=1.327$.

which defines $X_M = q_M/K_F$ and $X_C=q_C/K_F$. with $q_C=\sqrt{2}$ qm as 0.0938 and 0.1327 respectively. In the left side scale we set $\kappa=10^{-11}$ N, which produces values of 0.938 and 1.327 for q_M and q_C respectively. We note that R(q) is greater than zero for q in the range $0<q< q_C = \{[-f''-2\eta^2E/(1-\nu)]/2\kappa\}^{1/2}$; this means that only in that range will there be an increase in the fluctuaction of concentration and that these well defined values of wavelength will be characteristics of the spinodal decomposition. For values of $q>q_C$, R(q) is negative so the decomposition needs a mechanism different from the spinodal one. In this range of q, only systems of the metastable regime can undergo a tranformation.

In Figures 2 and 3 we show, for different aging times, the structure factor as a function of X for the linear and non-linear theories respectively. The upper and lower scales of $x=q/K_F$ correspond to different values of κ which produces a time shift in each plot. Also in each plot we show two initially different configurations: the dashed line corresponds to a constant A(q,0) equal to the value given by the gaussian distribution. The solid line correspons to an A(q,0) which depends upon q folllowing equation [10] and choosing $w=q_{M/5}$.

Figure 2 corresponds to the linear theory of Cahn. The aging time are 0, 10^3 and 10^4 for $\kappa=10^{-11}$ N, respectively. For $\kappa=10^{-9}$ N the aging times are 10^5 and 10^6 s. The time evolutions of $S^2(q,t)$ show a broadening towards small values of q_C, a higher maximum and an more violent cutoff of S^2 around q_C . The maximun of remains fixed in q_M and does not shift towards small values of q as has been determinated experimentally by Rundman *et al.* (24) and theoretically by Langer *et al.* (8).

Figure 3 corresponds to the non-linear theory of Tsakalakos (10), using the ap-

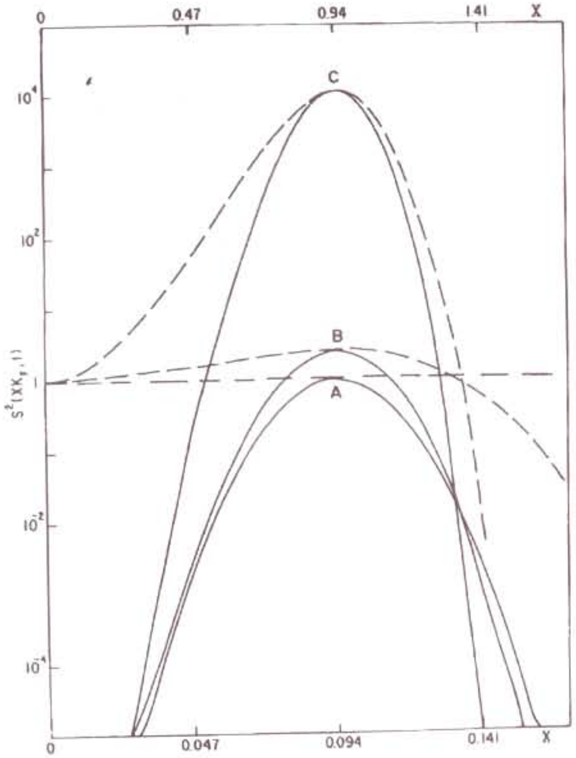

Figure 2. Structure factor S^2 versus X in the linear theory of Cahn. Upper scale for $\kappa=10^{-11}$. Lower scale for $\kappa=10^{-9}$ (------) $A(q,0) = 1.0$, (___) $A(q,0)$ defined by relation (10), choosing $w={}^{q}{}_{M}/5$. A: t=0 s. B: $t=10^3$ s or 10^5 s with $\kappa=10^{-11}$ or $\kappa=10^{-9}$, respectively.

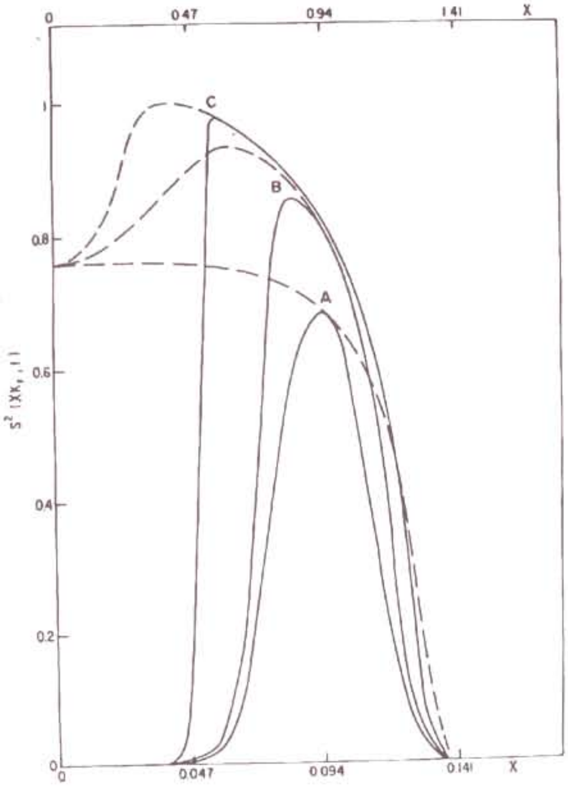

Figure 3. Same as Figure 2 in the non-linear theory of Tsakalakos.

proximate amplitude of Ditchek (22). In this Figure we use the same specifications as in Figure 2. It is worth noting the difference in the q dependence and in the evolution of S^2 as compared with the linear theory. The effect of the time t is to broaden S^2 toward the small values of q and the same time to shift the maximum of S^2 toward the same small q values. There is a tendency to qualitatively reproduce the time evolution of structure factor of Rundman *et al.* (24). The existence of the critical amplitude for finite time leads to coexistence of both the spinodal decomposition and the metastability; this is a characteristic of the non-linear theory. We note that the behaviour of S^2 in the limit of the decomposition ($A(q,t)=A_c$) coincides with the behaviour reported by Langer(6).

In Figure 4 we show the residual resistivity as a function of aging time for a process of spinodal decomposition. In Table 2 we specify the different conditions used for the calculations done using an Al-18%at.Zn alloy aged at 323 K.

In each curve of Figure 4, and independently of the conditions of the calculations, $\delta\rho(t)$ grows from $\rho(0)$ on during the whole process of decomposition. In the case of the linear theory there is not cutoff line for the growth, but in the non-linear theory, owing to the existence of the critical amplitude, $\delta\rho(t)$ tends towards a constant value,

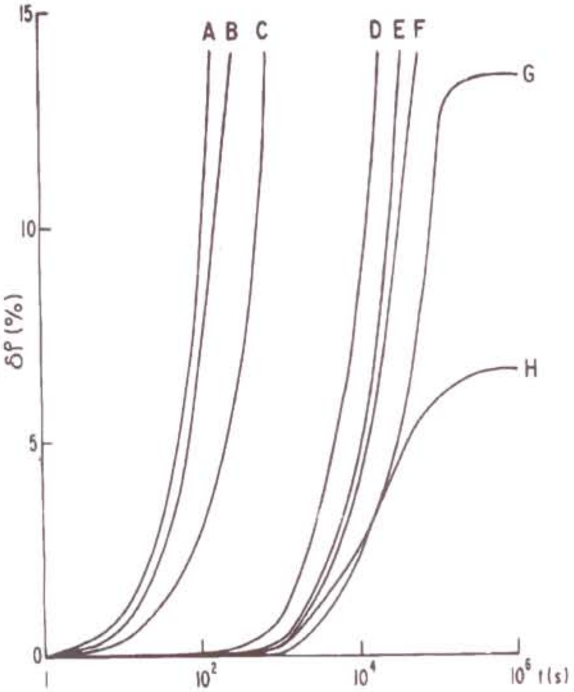

Figure 4. Variation of residual resistivity versus the aging time. Calculational specifications for A,B,C,D,E,F,G and H are given in Table 2.

as is showed by lines H and G. The effect of the different parameters given in Table 2 is to shift the time scale.

The behaviour of $\delta\rho$ which we have obtained is completely different from the one reported by Radomsky *et al.* (19) and slightly different from the one of Mimault *et al.* (18). If we take into account the experiments done in Al-Zn alloys at concentrations and temperature in the spinodal range, it is possible that part of the abnormal behaviour of the resistivity could be due the spinodal decomposition. In this sense we disagree with Mimault *et al.* (18), since our results do not lead us to attribute the total abnormal behaviour to the mechanism of spinodal decomposition, as they do.

Our results make us to think that the process of spinodal decomposition, both in the linear theory as well as in the non-linear one, is responsible for the monotonic growth of resistivity. In the non-linear case the limits of this growth are due to the existence of critical amplitude, which, structurally speaking, corresponds to the disappearence of the interconection which

Table 2

Specifications for calculations of Figure 4

Curve	κ(N)	a	w	Theory
A	10^{-11}	10^{-2}	$q_m/5$	Both
B	10^{-11}	1	*	Linear
C	10^{-11}	1	*	Non-linear
D	10^{-9}	1	$q_m/5$	Linear
E	10^{-9}	1	$q_m/5$	Non-linear
F	10^{-9}	1	*	Linear
G	10^{-9}	1	*	Non-linear
H	10^{-11}	1	$q_m/5$	Non-linear

*In these plots, $A(q,0) = a$

characterizes the spinodal decomposition. The second stage in the resistivity anomaly, that is, the decrease in resistivity, might be due to the mechanism of germination and growth, the drop of resistivity being a typical feature of growing of the GP zones (12).

Discussion

The behaviour of the resistivity reported in this work has, in both theories, an easy explanation, since under any parametric condition, the integration of [1] between 0 and q_c is grows the time. The $S^2(q,t)$ factor is always positive and greater than $S^2(q,0)$ (Figure 2 and 3). Also, $W^2(q)$ and q^3 are both positive. This explains the monotonic growth of $\delta\rho(t)$. In the non-linear case, for given time, the structure factor will be bounded by the critical amplitude, thus $\delta\rho$ tends to show a plateau.

If we consider q values between q_c and $2K_F$, $S^2(q,t)$ will continue to be positive, but less than $S^2(q,0)$, as it is show in Figure 2 for linear case. For the non-linear case this is not evident but it will be enough to check the results of Binder *et al.* (27) or those of Langer *et al.* (8) to be sure that this condition also holds. This factor give the chance that $\delta\rho(t)$ may become negative, which will depend only upon the thermodynamic parameters used. In the case that q_M were located in the limit of small angles, the integration between 0 and 2 K_F will lead to a monotonic decrease of $\delta\rho$ similar to the one obtained by Radomsky *et al.* (19). It is clear that values of q greater than q_c lead to a decrease of fluctuation of the concentration, so that, in this stage, the mechanism which causes the decomposition is different from the spinodal one. We think this is particularly the case of Radomsky *et al.* (19), since their relation [6] establishes as limits of integration the value $0 \leq 2 \leq K_F$. In the case that q_M lies in the limit of large q_c, the positive contribution of relation [12] between 0 and q_c dominates the negative contribution between q_c and $2K_F$.

The use of the values suggested by Radomsky *et al.* (19) or Rundman *et al.* (24) for calculational parameters does not modify the behaviour of $\delta\rho(t)$ reported in this work.

The influence of the parameters a, w and κ do not determe the form and dependence of $\delta\rho$ upon time. Fixing w and some other thermodynamic parameters, a does not modify the value of $\delta\rho$ calculated using the linear theory; however, it does modify it in the non-linear theory in such way that evolution is faster the smaller a is set. The consideration or not of the dependence of A(q,0) upon q does not seem to be an important matter in either of the two theories. Following the work of Huston *et al.* (26) we can show that A(q,0) is proportional to $C_o(1-C_o)$ as proposed by Guinier *et al.* (28). The effect of κ is to shift the time scale. A fitting of the plots obtained in this work with experimental plots could lead us to more realistic values of these parameters.

Finally as a consecuence of our results, we can answer the question which names this work:

The process of spinodal decomposition in the linear and non-linear theories considered in this work is not able to explain by itself the anomaly of resistivity, since during that process $\delta\rho$ grows at the same rate that the fluctuation of concentration grows. In the non-linear case there is a bound in the grows due to the existence of a critical amplitude. The decrease of the $\delta\rho$ observed under some conditions can be associated with an increase of the Guinier-Preston zones. In this way, the resistivity anomaly could be explained by using a combination of the process of spinodal decomposition and the process of germination and growth, one of them explaining the increase and the process explainig the decrease of $\delta\rho$.

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