On the Behavior of the Lifshitz Line in Ternary Homopolymer/Diblock-Copolymer Blends

Alexander Kudlay*1,2 and Semjon Stepanow1
1Fachbereich Physik, Martin-Luther-Universität Halle, D-06099
Halle, Germany
2Physics Department, Moscow State University, 117234 Moscow, Russia

Abstract

We present results of the study of the non-monotonous behavior of the Lifshitz line as a function of temperature in ternary homopolymer/diblock-copolymer mixtures. The non-monotonous behavior of the Lifshitz line is due to the wave vector dependence of fluctuational corrections, which we treat in the framework of the renormalization group method. Our results are in agreement with the experimental findings of Schwahn et al. [1,2].

I. INTRODUCTION

The Lifshitz point appears in systems with competing tendencies for phase separation into bulk or spatially modulated phases. If the appropriate parameter controlling the relative strength of the two tendencies is varied along the critical line of phase transitions a special multicritical point occurs at which the character of phase separation undergoes a change from bulk phase separation to the phase separation into a spatially modulated phase. The Lifshitz point is known to exist in magnetic systems [3]-[5], liquid crystals [6], polyelectrolytes [7]-[8], oil/water/surfactant mixtures [9], random block-copolymers [10]-[11], mixtures of homopolymers and diblock copolymers [12,13]. First theoretical interest in such systems was generated by the work of Hornreich et al. [4], who introduced the Lifshitz point and calculated the critical exponents for this class of universality. Most of the theoretical effort since has been concentrated on calculating the values of the exponents via application of various renormalization group techniques [14].

The aim of the present work is the theoretical description of the behavior of the Lifshitz line with varying temperature. Recent experiments [1,2,15,16] on symmetric isoplethic A/B/A-B homopolymer/diblock-copolymer mixtures in the vicinity of the Lifshitz conditions revealed many challenging phenomena not accounted for by the existing mean-field theories [2,3]. In particular, one of the experimental results in clear disagreement with the mean-field prediction is the non-monotonous behavior of the Lifshitz line [1,2] with temperature. We will show that the wave vector dependence of the fluctuation corrections is responsible for the experimentally observable shift of the Lifshitz line from its mean-field value. The fluctuation effects will be taken into account within the one-loop renormalization group method.
We will put special emphasis on the comparison between the theoretically predicted behavior of the Lifshitz line and current experimental results. As we will demonstrate, a major factor determining the character of this behavior is the value of the lower critical dimension $d_l$, a fact which has been little discussed in the literature. It is important to stress that the actual renormalized value of $d_l$ is not known at present. Therefore, since the mean field value $d_{mf} = 4$ is close to $d = 3$ — dimension of space of the considered polymer blends, we will theoretically analyze different types of behavior of the Lifshitz line resulting from different possible values of $d_l$ and compare them with the experiment.

II. PERTURBATIVE CALCULATION OF THE SHIFT OF THE LIFSHITZ LINE

We start from the conventional expansion of the Landau free energy functional

$$H[\psi(q)] = \frac{1}{2} \int_q \psi(-q) G_0^{-1}(q) \psi(q) + \frac{\lambda}{4!} \int_q \int_q \int_q \int_q \psi(q_1) \psi(q_2) \psi(q_3) \psi(-q_1 - q_2 - q_3)$$

in powers of the Fourier transform of the order parameter. In particular, for the ternary mixtures under consideration the natural order parameter is the deviation (from the volume averaged) of the concentration of $a$ (or $b$, since the system is symmetric) monomers. The parameters of the Hamiltonian (1) for the polymer system can be obtained from coarse-graining of the corresponding microscopic Hamiltonian. In particular, near the Lifshitz line (to be defined later) the bare correlation function $G_0^{-1}(q)$ can be written as follows:

$$G_0^{-1}(q) = \tau + c_1(\phi) q^2 + c_2 q^4$$

with $\tau \sim (T - T_c)/T$ being the reduced temperature. For the considered ternary mixtures the coefficient $c_1(\phi)$ depends on the concentration of diblock-copolymer $\phi$ and changes the sign with the variation of $\phi$. Within the mean-field theory the Lifshitz point is defined by the two conditions: $\tau = 0$ and $c_1(\phi_{MFLP}) = 0$. More generally, we can introduce the mean-field Lifshitz line as the locus of points in parameter space $(\phi, \tau)$ at which the quadratic term vanishes: $c_1(\phi_{MFL}(\tau)) = 0$. The Lifshitz line (LL) is easily determined experimentally by considering the position of the peak of the static scattering curve. If we begin increasing the concentration of diblock at constant temperature the LL is determined by the diblock concentration at which the peak in the scattering curve first shifts off the zero wave vector. A noteworthy feature of the considered polymeric system is that the temperature $T$ enters the Hamiltonian (1) only via the Flory-Huggins parameter, hence the coefficient $c_1(\phi)$ turns out to be independent of temperature, so that the mean-field position of the LL $\phi_{MFLLL}$ is also temperature independent and is determined solely by the ratio of the molecular weights of the polymers comprising the mixture. This mean-field prediction was not confirmed experimentally. Instead of being constant, the position of the LL was found to vary with temperature, more precisely, it exhibited a non-monotonous behavior, which shows that fluctuations should be taken into account. This is hardly surprising because at the LL, when $c_1(\phi)$ vanishes, the fluctuation corrections in fact become the only input into the renormalized counterpart of $c_1$ and thus always play a role.

Let us consider the renormalized correlation function. Note, that since our goal is to calculate the deviation of the LL, the renormalized quadratic term vanishes:
\[ G^{-1}(q) = \tau + l_1(\phi, \tau) q^2 + c_2 q^4 \]  

(3)

\[ l_1(\phi, \tau) = c_1(\phi) + \Delta c_1(\tau) = 0 \]  

(4)

The shift of the LL (which is temperature dependent due to fluctuation corrections) is denoted by \( \Delta c_1(\tau) \). Within the approximation we use in this paper \( c_2 \) will not be renormalized. The renormalized parameter \( l_1 \) can be found by considering the Dyson equation:

\[ G^{-1}(q) = G_0^{-1}(q) - \Sigma(q), \]  

\[ \Sigma(q) = D_1(q) + D_2(q). \]  

(5)

(6)

We consider in the self-energy \( \Sigma(q) \) only the one and two-loop diagrams:

\[ D_1 = -\frac{n+2}{6} \lambda \int \frac{1}{q \tau + c_2 q^4} \]  

(7)

\[ D_2(q) = \frac{\lambda^2}{6} \int_{q_1} \int_{q_2} \frac{1}{[\tau + c_2 q_1^4][\tau + c_2 q_2^4][\tau + c_2 (q_1 + q_2 + q)^4]} \]  

(8)

For generality and ease of comparison with known results we have introduced \( n \) — the number of components of the order parameter. Note, that for the polymer blends [1,2], whose description is the goal of our work, due to the incompressibility condition the order parameter is a scalar, i.e. \( n = 1 \), as is indeed clear from the Hamiltonian [1]. The \( D_1 \) diagram is \( q \)-independent and is therefore of no relevance to the renormalization of \( c_1 \). The first correction to it is given by \( D_2(q) \). Calculation of \( D_2(q) \) is performed easier in the real space. For the experimentally relevant case \( d = 3 \) we use the \( r \)-space representation of the correlation function

\[ G(r) = \int \frac{d^3 q}{(2\pi)^3} \frac{\exp(iqr)}{\tau + c_2 q^4} = \frac{\xi}{4\pi c_2 r/\xi} \frac{1}{\exp(-r/(\xi \sqrt{2}))} \sin \left( r/(\xi \sqrt{2}) \right) \]  

(9)

to rewrite the expression for \( D_2(q) \) in terms of \( G(r) \) as follows:

\[ D_2(q) = \lambda^2 \int e^{iqr} G^3(r) d^3 r \]  

(10)

where \( \xi = (c_2/\tau)^{1/4} \) is the mean-field correlation length. In fact we need only the quadratic term of the diagram in powers of \( q \), which is readily calculated:

\[ D_2^{(2)}(q) = -b \frac{\lambda^2 \xi^8}{c_2^3} q^2 \]  

(11)

where \( b \approx 0.109 \times 10^{-4} \) is a constant. As it is clear from the Dyson equation [1] this result gives in fact the shift of the LL:

\[ \Delta c_1(\tau) = B \frac{\lambda^2 \xi^8}{c_2^3} \]  

(12)

Note, that we have obtained the expression [12] within the perturbation theory and therefore it is not valid in the regime of strong fluctuations. However, the scaling behavior of \( \Delta c_1(\tau) \) in
the regime of strong fluctuations can be obtained from (12) by replacing $\lambda$ with the effective coupling constant $\lambda_r$, and understanding under $\xi$ the true correlation length, thus

$$\Delta c_1(\tau) \simeq B\frac{\lambda_r^2(\tau)\xi^8(\tau)}{c_2^2}. \quad (13)$$

This formula immediately allows some conclusions about the qualitative behavior of the LL.

First of all, we observe that the correction is positive, which means that on the LL $c_1(\phi) < 0$, i.e. fluctuations shift the LL into the $q_* \neq 0$ region of the mean-field theory. For the homopolymer/diblock copolymer blend that means that LL shifts to greater concentration of diblock, which is in agreement with experiments [15,16,1,2]. Next consider the dependence on temperature. We have two regimes here: perturbative (small correlation lengths, at high temperatures) and scaling (low temperatures, large $\xi$). In the perturbative regime where the input of fluctuations is small, $\lambda$ remains practically non-renormalized, so that with lowering temperature $\Delta c_1(\tau)$ should increase simply due to the increase of $\xi$. In the scaling regime the main effect (as will be shown below) comes from the renormalization of the coupling constant $\lambda_r$. In fact in this regime we can obtain the scaling dependence of the correction from the considerations of dimensionality: demanding that the $\Delta c_1(\tau)q^2$ term of the Hamiltonian (3) have the same dimensionality in $\xi$ as the $c_2q^4$ term of the correlation function. If $c_2$ is not renormalized (as is our case), then

$$\Delta c_1(\tau) \sim \xi^{-2} \quad (14)$$

As we can see, in the scaling regime the correction decreases with increasing $\xi$ (i.e. decreasing temperature). Combined with the conclusion made above about the increase of $\Delta c_1(\tau)$ in the perturbative regime we come to conclusion that $\Delta c_1(\tau)$ exhibits a non-monotonous behavior as a function of $\tau$. This behavior is a manifestation of the crossover between the regimes of small and strong fluctuations.

III. RENORMALIZATION GROUP STUDY OF THE LIFSHITZ LINE

To describe $\Delta c_1(\tau)$ quantitatively we have to obtain expressions for $\lambda_r(\tau)$ and $\xi(\tau)$ in both the perturbative and scaling regimes. For this purpose we shall employ a renormalization group technique to the first order in $\varepsilon$ (one-loop RG). Note, that within this method the parameter $c_2$ does not renormalize. The renormalization of temperature is described by the one-loop diagram (7):

$$D_1 = D_1^0 + D_1' = -\frac{n+2}{6}\lambda\int_q \frac{1}{c_2(q^2)^2} + \frac{n+2}{6}\tau\lambda\int_q \frac{1}{c_2(q^2)^2(\tau + c_2(q^2)^2)}. \quad (15)$$

The above expression is conventionally split into two parts responsible for additive and multiplicative renormalization of temperature. For dimensions $d > 4$ a cutoff at the upper limit in integration over $q$ in the first term is assumed. These two terms give the critical dimensions of the Lifshitz class of universality. The lower critical dimension $d_l$ is defined as the dimension when the first term logarithmically diverges at small $q$. The upper critical dimension $d_u$ is the dimension at which the second term logarithmically diverges at small $q$ for zero temperature. A cutoff at the lower limit of integration over $q$ is implied in Equation
For the isotropic Lifshitz class of universality we obtain: $d_{l}^{mf} = 4$ and $d_{u}^{mf} = 8$. The real experimental system corresponds to $d = 3$ so that we come to conclusion that we are situated below $d_{l}^{mf}$. This means that the $D_{a}^{q}$ term diverges at small $q$, i.e. for large correlation lengths and thus no phase transition of the second order is possible at a finite temperature. However, this value for the lower critical dimension is only the mean-field one. Fluctuations renormalize the value of the lower critical dimension. The calculation of the renormalized lower critical dimension is a formidable task, so that the true renormalized value of $d_{l}$ for the experimental system is actually unknown. Therefore, we will consider below several possibilities.

Going back to renormalization of $\tau$ by substituting (15) into the Dyson equation we obtain:

$$\tau_{r} = \tau_{a} Z_{2}(\Lambda_{\text{min}})$$

(16)

$$\tau_{a} = \tau - D_{1}^{a} Z_{2}(\Lambda_{\text{min}})^{-1},$$

(17)

where for purposes of clarity we introduced the temperature with additive term $\tau_{a}$ as well as renormalized temperature $\tau_{r}$. The quantity $\Lambda_{\text{min}}$ in Equation (16-17) is the lower cutoff imposed in Equation (15) in integration over the momentum $q$. The RG treatment is based on the following perturbative expression:

$$\tau_{r} = \tau_{a}\left(1 - \frac{n + 2}{6}\lambda \int_{q} \frac{1}{c_{2}(q^{2})(\tau + c_{2}(q^{2}))^{2}} + \ldots\right)$$

(18)

Likewise considering the fluctuation correction to $\lambda$ for its renormalized counterpart $\lambda_{r}$ we obtain:

$$\lambda_{r} = \lambda\left(1 - \frac{n + 8}{6}\lambda \int_{q} \frac{1}{(\tau + c_{2}(q^{2}))^{2}} + \ldots\right)$$

(19)

These two equations are the starting point to derive the differential equations of the renormalization group for $\tau_{r}$ and $\lambda_{r}$. To do this we introduce a running cutoff $\Lambda$ at the lower limit of the integrals in (18)-(19), differentiate both parts of these equations with respect to $\Lambda$ and replace in the rhs the bare quantities $\tau$ and $\lambda$ through the effective ones. Thus we obtain:

$$\Lambda \frac{\partial \ln \tau_{r}}{\partial \Lambda} = \frac{n + 2}{6} g$$

(20)

$$\Lambda \frac{\partial g}{\partial \Lambda} = -\varepsilon g + \frac{n + 8}{6} g^{2}$$

(21)

where the effective dimensionless coupling constant is defined as follows $g = \bar{\lambda}_{r} \Lambda^{-\varepsilon}$ with $\varepsilon = 8 - d$, and $\bar{\lambda} = \lambda K_{d}/c_{2}^{2}$, $K_{d} = S_{d}/(2\pi)^{d}$, $S_{d}$ being the surface of a unit $d$-dimensional sphere. As can be seen from (21) the fixed point of the effective coupling constant, $g = \bar{\lambda}_{r} \Lambda_{\text{min}}^{-\varepsilon}$, is $g^{*} = \frac{6}{n + 8} \varepsilon$.

Notice that the second equation is independent of $\tau$, therefore we solve it first and then substitute the result $g(\Lambda)$ into the first one. Thus we obtain the solution of the differential equations (20)-(21):
\[ \tau_r = Z_2(\Lambda_{\text{min}}) = \left(1 + \frac{n + 8 \bar{\lambda}}{6 \varepsilon} \Lambda_{\text{min}}^{-\varepsilon}\right)^{-\frac{n+2}{n+8}} \] (22)

\[ \frac{\lambda_r}{\lambda} = \left(1 + \frac{n + 8 \bar{\lambda}}{6 \varepsilon} \Lambda_{\text{min}}^{-\varepsilon}\right)^{-1} \] (23)

Equation (22) for \( \tau_r \) allows us to obtain the critical exponent of the correlation length: \( \nu = \frac{1}{4} \left(1 + \frac{n + 8}{n + 8} \varepsilon\right) \), which is a well-known result [4,5]. Note, that in this system \( \tau_r \) is expressed via the temperature with additive shift (RG generalization of Equation (17)):

\[ \tau_a = \tau + \frac{n + 2}{6} \int_q \frac{\lambda_r(q)}{Z_2(q)c_2q^4} \] (24)

Taking into account the renormalization of the coupling constant \( \lambda_r \) and the propagator in the expression of the shift of the critical temperature can be found by considering the higher-order corrections to the self-energy \( \Sigma(q) \). It is clear that the infrared behavior of these corrections is controlled by the momentum \( q \), which is the argument of the self-energy \( \Sigma(q) \). This demands to write \( \lambda_r(q) \) and \( Z_2(q) \) under the integral in (24) as functions of the external momentum \( q \). The relation (24) makes the one-loop RG scheme for renormalization of \( \tau_r \) and \( \lambda_r \) complete. Now, using the relation between \( \Lambda_{\text{min}} \) and \( \xi \) (see below) we can obtain from Equation (22) \( \xi(\tau) \), which substituted into (23) will give \( \lambda_r(\tau) \). The two dependences substituted in turn into the formula for \( \Delta c_1(\tau) \) will give our final result — the deviation of the LL from the mean-field value as a function of temperature. In order to find relation between the cutoff wave vector \( \Lambda_{\text{min}} \) and the correlation length \( \xi \), one should find the perturbative limit of the RG formulae (22) or (23) and demand it to be equal to the corresponding diagrams (18) or (19). Thus it is straightforward to obtain: \( \Lambda_{\text{min}} = \xi^{-1} \).

Using this relation and introducing reduced variables we can rewrite Equation (22)-(24) as follows:

\[ \tilde{\tau}_r = Z_2(\xi) = \left(1 + \bar{\lambda} \xi \varepsilon\right)^{\frac{n+2}{n+8}}, \quad \tilde{\tau}_a = \tilde{\tau} + a \int_{\xi^{-1}}^{\infty} \frac{\tilde{\lambda}_r(q)}{Z_2(q)c_2q^d} dq \] (25)

\[ \frac{\bar{\lambda}_r}{\lambda} = \left(1 + \bar{\lambda} \xi \varepsilon\right)^{-1} \] (26)

where the reduced variables are: \( \bar{\lambda} \equiv \frac{n+8}{6 \varepsilon} \bar{\lambda}, \tilde{\tau} \equiv \frac{\tau}{c_2}, \) and the constant \( a \equiv \frac{n+2}{n+8} \varepsilon \). However the relation \( \Lambda_{\text{min}} = \xi^{-1} \) is asymptotically correct only in the vicinity of the upper critical dimension \( d_u = 8 \) and we do not expect it to hold for the considered experimental situation \( d = 3 \). Therefore, we only know that \( \Lambda_{\text{min}} \sim \xi^{-1} \) with the prefactor being unknown. In this situation the constants \( a \) and a new constant \( f \) (defined via \( \bar{\lambda} \equiv f \lambda/c_2^2 \)) become essentially fit parameters of the theory. The expression for \( \Delta c_1(\tau) \) in reduced variables reads:

\[ \frac{\Delta c_1(\tau)}{c_2bf^{-2}} = \tilde{\lambda}_r^2(\tilde{\tau})\xi^8(\tilde{\tau}) \] (27)

Before solving the system let us make some further comments on the Equation (22)-(24), in particular discuss the issue of the lower critical dimension. To that end we should consider the scaling of the term responsible for additive renormalization:
\[
\Delta \tau \equiv a \int_{\xi^{-1}}^{\infty} \frac{\lambda_r(q)}{Z_2(q)l_2(q)q^d q^{d-1} dq} \sim \xi^{-1/\nu}
\] (28)

If \( d > d_l \) then this integral converges on small \( q \) and this addition to temperature can be neglected in the RG calculations as it is does not depend on the correlation length. Note, however that experimentally this correction is still relevant. Since the values of the fluctuation shift of temperature are different in the Ising and Lifshitz classes of universality the temperature of the LP is shifted to lower values then the transition temperature of the Ising class of universality. The situation of the lower critical dimension corresponds to logarithmic divergence of \( \Delta \tau \), i.e. \( 1/\nu = 0 \). If \( d < d_l \) then the correction diverges at \( \xi \to \infty \), which precludes the phase transition at finite temperatures, the transition temperature goes to zero. According to Equation (28) we can consider the exponent \( \nu \) in this case to be formally negative. As we have mentioned above the renormalized value of \( d_l \) is not presently known, so we will consider the two possibilities: \( d > d_l \) and \( d < d_l \). Therefore, it does not make sense to use the exponent \( \nu \) obtained to order \( \varepsilon \), since we do not expect it to be correct for \( d = 3 \). On the contrary, using the scaling relations (supposed to be correct even for negative \( \nu \)) we will express the exponents in the equation for \( \xi(\tilde{\tau}) \) (29) via the exponent \( \nu \) (generally the exponent \( \eta \) is also necessary, however it is zero in the one-loop approximation). Thus, we arrive at the equation

\[
\left( \tilde{\tau} + a\tilde{\lambda} \int_{\xi^{-1}}^{\infty} \left( 1 + \tilde{\lambda} q^{-\varepsilon} \right)^{1+4(1-1/\nu)/\varepsilon} \frac{q^{d-1} dq}{q^{d}} \right)^{-1} = \xi^4 \left( 1 + \tilde{\lambda} \xi^{\varepsilon} \right)^{-4(1/\nu)/\varepsilon}
\] (29)

Its solution \( \xi(\tilde{\tau}) \) substituted into (29) gives \( \tilde{\lambda}_r(\tilde{\tau}) \) and thus we can obtain the shift of the LL \( \Delta c_1(\tilde{\tau}) \) according to (27).

The results of numerical evaluation of \( \Delta c_1(\tilde{\tau}) \) according to Equation (27-29) are plotted in Figure 1. We have considered two cases: \( d > d_l, \nu = 1 \) — solid line (\( \tilde{\lambda} = 1, a = 2 \)); \( d < d_l, \nu = -1 \) — two dashed curves (\( \tilde{\lambda} = 1 \) for both curves; \( a = 0.5, a = 2 \)). If \( d > d_l \) then for all values of parameters \( a \) and \( \tilde{\lambda} \) the LL has the qualitative form as the curve plotted in Figure 1: with decreasing temperature \( \Delta c_1(\tilde{\tau}) \) initially increases (due to increase of \( \xi \) as explained above), then for large fluctuations decreases (due to decrease of the renormalized coupling constant \( \lambda_r \)) and the LL ends at a Lifshitz point. Note that the LP occurs at the negative temperatures \( \tilde{\tau} \) due to the finite shift of transition temperature discussed above. For \( d < d_l \) the situation is more complex. At large values of \( a \) (a = 2 curve of Figure 1) the LL goes to \( \tilde{\tau} \to -\infty \), so that no Lifshitz point exists (we may say it is shifted to \( \tilde{\tau} = -\infty \)). As can be seen from Figure 1 in other respects the LL of this case is qualitatively the same as in the case \( d > d_l \). At small values of \( a \) (a = 0.5 curve of Figure 1) two solutions of Equation (29) exist at large temperatures (the one corresponding to greater \( \xi \) is plotted with the dot curve in Figure 1) and no solution at small temperatures. Thus in this case the LL ends in a point at which the correlation length is finite, which indicates that this is a point of the phase transition of the first order. It is not clear however, whether this case represents physical features or is an artifact of approximations of the theory.

Comparing obtained results with the experimental data of Schwahn et al. [11,2] we observe that the experimental LL has the same qualitative shape. We would like to stress that the present study predicts that the Lifshitz line approaches its mean-field counterpart for high and deep temperatures, the prediction which is also in agreement with experimental behavior.
of the Lifshitz line found in [1,2]. However, on the basis of the current data of Ref. [1,2] it is not clear which of three cases concerning the value of the lower critical dimension of the system which we have discussed above, actually takes place.

Summarizing, we have calculated the deviation of the LL from the mean-field behavior by taking into account the first fluctuational correction to the $c_1$ coefficient. The renormalized properties are calculated within the one-loop renormalization group. Our calculations are carried out in the immediate vicinity of the Lifshitz line, so that no crossover to either Ising or Brazovsky universality class is considered. The obtained behavior of the LL qualitatively agrees with that observed in the scattering experiments on ternary homopolymer/diblock-copolymer systems. The approach we have presented in the present article is based on the Landau-Ginzburg expansion of the free energy with phenomenological parameters $\lambda$, $c_1$, $c_2$. Of course, this fact restricts a complete quantitative comparison with experiment. However, we point out that despite this the agreement of the behavior of the computed Lifshitz line at high and low temperatures with experimental one is a strong support of the validity of our approach. The major reason for the uncertainty in the complete quantitative comparison with the experiment is due to unknown values of the critical exponents of the Lifshitz universality class. Depending on the value of the lower critical dimension in the renormalized theory the LP exists or it is shifted to infinitely small temperatures. More experiments are needed to determine which of the possibilities actually takes place, which would amount to experimental determination of the lower critical dimension of the system. We hope that this work will stimulate such investigations.

The authors acknowledge stimulating discussions with E. Straube, I. Erukhimovich, and D. Schwahn. A. K. acknowledges support of the Graduirtenkolleg “Polymerwissenschaften”.

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Figure 1. The shift of the Lifshitz line $\Delta c_1$ as a function of temperature $\tilde{\tau}$. Solid curve: $d > d_l$, dash curves: $d < d_l$. For all curves $\lambda = 1$. 
Fig. 1

A. Kudlay and S. Stepanow