PHYSICAL REVIEW B VOLUME 21, NUMBER 10 15 MAY 1980

Effect of the substrate potential on incommensurate epitaxies at finite temperatures

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(Received 5 November 1979)

Incommensurate epitaxial systems at $T = 0$ were first studied by Frank and Van der Merwe. Here we investigate these systems at finite temperatures. We suggest that thermal fluctuations can reduce the effect of the substrate potential sufficiently that at some finite temperature the modulation of the epitaxy due to the substrate is qualitatively changed from the behavior near $T = 0$.

I. INTRODUCTION

The effect of a periodic potential on an incommensurate epitaxy has received a great deal of attention lately in a number of different contexts: in the study of physisorption of monolayer rare gases on graphite,[1] in surface reconstruction,[2] in superionic conductors,[3] in the layered chalcogenides,[4] and in the "one-dimensional" organometallic compounds.[5] These studies basically focus on a model first studied by Frank and Van der Merwe[1] in the context of epitaxial growth, which is described by the Hamiltonian

$$\begin{aligned} H &= H_x + H_y\,, \\ H_x &= \frac{\mathcal{J}}{3}\left(\sum_{i,\delta_x}(x_i - x_{i+\delta_x} - a)^2 + \sum_{i,\delta_y}(x_i - x_{i+\delta_y})^2\right) \\ &\quad + \lambda\sum_i \cos\frac{2\pi x_i}{b}\,. \end{aligned} \tag{1}$$

Here x_i denotes the x coordinate of an epitaxial atom at site i and $\delta_{x(y)}$ is a nearest-neighbor vector in the $x(y)$ direction. a, the "natural" epitaxial atomic spacing, is in general different from the substrate atomic spacing b. $\mathcal{J}$ is a kind of elastic coefficient of the epitaxial atoms, while λ measures the interaction strength between the epitaxy and the substrate. In general, the epitaxial Hamiltonian should also contain cross terms involving both the x and the y coordinates of the epitaxial atoms, but in this simple model the y coordinate is assumed decoupled from the x coordinate, and H_y need not be independently investigated.

The ground-state configuration of H has been discussed by Frank and Van der Merwe.[1] For a not too different from b, the epitaxy remains commensurate with the substrate; the equilibrium lattice spacing of the epitaxy is still b. When the difference of a and b exceeds a certain value, the epitaxy becomes incommensurate with the lattice and misfit dislocations are formed continuously.

For some applications where large thermal fluctuations are present (for which two-dimensional systems are notorious), the finite-temperature situation has to be investigated separately. The commensurate-incommensurate transition (CIT) at nonzero temperatures has recently been considered by Pokrovskii and Talapov[6] and by Bak, Mukamel, Villain, and Wentowska[7] theoretically and by Chinn and Fain,[8] Stephens, Heiney, Birgeneau, and Horn,[9] and Vora, Sinha, and Crawford[10] experimentally. The CIT, however, is not the focus of the present paper. We are concerned with the properties of the incommensurate system only. We suggest that there can be at least two different incommensurate regions: (1) a low-temperature phase where the substrate potential is strong enough to produce significant misfit modulations, and (2) a high-temperature phase where thermal fluctuations reduce the effect of the substrate potential so much that the modulation of the epitaxy is qualitatively changed. We predict that there is an infinite response for the epitaxy at a wave vector near $2\pi/a - 2\pi/b$ in the low-temperature region, whereas in the high-temperature region this response is finite.

These results are based on perturbation calculations from both the low- and the high-temperature sides. These will be reported in Sec. II and III. The implications of our results are discussed in Sec. IV.

II. THE LOW-TEMPERATURE REGION

We assume a is sufficiently different from b that the incommensurate phase is stable at $T = 0$. In this case, one can expand the atomic coordinates x_i in a Fourier series, as first discussed by Ying[1] and more recently by McMillan,[4] Shiba,[11] and Theodorou and Rice.[12] Close to the CIT, many Fourier components have to be kept, and the original treatment of Frank and Van der Merwe in terms of misfit dislocation is simpler. Far away from the CIT, however, the misfit dislocations overlap and a description in terms of a Fourier series is a more appropriate choice. We shall assume that the system is sufficiently far from the CIT so that we need keep only the first term of the Fourier series. In the earlier studies,[1,12] this turned out to be a good approximation over a wide range of parameters. Under these approximations the calculation of Ying can be easily generalized to nonzero temperatures.

A simple physical picture of the zero-temperature situation is the following. In order to take advantage of the substrate, the epitaxy will exhibit a periodic modulation and the average interatomic spacing will achieve some value c in between b and a. At low temperatures we expect the atoms to undergo small vibrations about their modulated equilibrium positions. Thus we write

$$x_i = i_x c + \delta x_i + t \sin[2\pi(i_x c + \delta x_i)/b] , \qquad (2)$$

where c is the average spacing of the incommensurate epitaxy, δx_i is the deviation from this equilibrium spacing, and t is a parameter determined from the equilibrium condition. Note the appearance of δx_i inside the sine function. This occurs because the incommensurate phase can slide freely over the substrate[1] and in general $\langle \delta x_i^2 \rangle = \infty$. Thus δx_i itself is not a small expansion parameter; the proper small parameter is actually $\delta x_i - \delta x_{i+\delta_x}$ (or $\delta x_i - \delta x_{i+\delta_y}$), the *difference* in positions of nearest-neighbor atoms.

There are many ways by which the low-temperature calculation can be carried out. In order to connect with later work on dynamics, we chose to study here the static limit of the appropriate Langevin equation. Differentiating Eq. (1), we thus write

$$\partial_t x_i = \mathcal{J} \Delta^2 x_i + \eta_i + \Delta\mu_i + \lambda \sin(2\pi x_i/b) . \qquad (3)$$

Here η is a thermal white noise whose second moment is given by $\langle \eta_i(t) \eta_j(t') \rangle = 2\Gamma \delta_{ij} \delta(t-t')$ with Γ the kinetic coefficient. $\Delta\mu_i$ is a small external driving field introduced to facilitate the calculation of linear-response coefficients, and Δ^2 is the usual discrete double-difference symbol. The method of calculation given below is very similar to Ying's $T=0$ work. We reproduced it here only to point out what approximations are valid at finite temperatures.

Our main focus will be the structure factor $S_{\vec{q}} = \sum \langle \exp[i\vec{q} \cdot (\vec{r} - \vec{r}')] \rangle$. First let us develop some preliminaries. Based on our physical argument given above, we substitute a trial solution of the form of Eq. (2) in Eq. (3) and get

$$\begin{aligned}\partial_t x_i = {} & \mathcal{J} \Delta^2 \delta x_i (1 + t \cos[(2\pi/b)(c i_x + \delta x_i)]) + \eta_i + \Delta\mu_i + \{t \sin^2(\pi c/b) + \lambda J_0(t)\} \sin[(2\pi/b)(c i_x + \delta x_i)] \\ & + \lambda 2 J_1(2\pi t) \cos[(2\pi/b)(c i_x + \delta x_i)] \sin[(2\pi/b)(c i_x + \delta x_i)] + \cdots . \end{aligned} \qquad (4)$$

We have used the fact that

$$\Delta^2 \sin[(2\pi/b)(c i_x + \delta x_i)] \cong \Delta^2(\delta x_i)(2\pi/b) \cos[(2\pi/b)(c i_x + \delta x_i)] + \sin^2(\pi c/b) \sin[(2\pi/b)(c i_x + \delta x_i)] + O((\Delta^2 \delta x_i)^2) \qquad (5)$$

and

$$\begin{aligned} \cos(x \sin y) &= J_0(x) + \sum_{s=1}^{\infty} J_{2s}(x) \cos 2sy , \\ \sin(x \sin y) &= 2 \sum_{s=0}^{\infty} J_{2s+1}(x) \sin(2s+1)y . \end{aligned} \qquad (6)$$

Equation (6) is just a mathematical expansion in terms of the Bessel function J_s (not to be confused with the elastic constant $\mathcal{J}$). Note that it is in Eq. (5) that the smallness of the parameter $\Delta^2 \delta x_i$ is used. In physical problems of interest, $\lambda/\mathcal{J}$, and hence $t/\mathcal{J}$, is a small number.[13] Hence the last term of Eq. (4) is indeed smaller than the rest of the terms. It is kept only to provide the reader an idea of the perturbation scheme involved. For time-independent phenomena $\partial_t x_i = 0$ and the coefficient in the curly bracket in Eq. (4) must be equal to zero,

$$t \mathcal{J} \sin^2(\pi c/b) + \lambda J_0(t) = 0 . \qquad (7)$$

This is essentially the same as Ying's[1] result [see his equations (5)–(8)]. From the rest of Eq. (5) we get

$$\mathcal{J} \Delta^2 \delta x_i \{1 + t \cos[(2\pi/b)(c i_x + \delta x_i)]\} + \eta_i + \Delta\mu_i = 0 . \qquad (8)$$

From this, δx_i can be determined in terms of η and $\Delta\mu$. The solution of (8) is basically one of determining the phonon dispersion of the incommensurate phase and has been discussed recently by many authors.[1,12,14] It was found that one could expand δx_i as a Fourier series:

$$\delta x_i = \sum_q \delta x_q \exp(i\vec{q} \cdot \vec{r}_i) . \qquad (9)$$

For q far away from $2\pi(c-b)/b^2$, the phonon dispersion is only weakly modified by the "incommensurate" term $t \cos[(2\pi/b)(cix + \delta x_i)]$ so that

$$\begin{aligned} \delta x_q &\simeq (\eta_q + \Delta\mu_q)/(\mathcal{J} \alpha_q) , \\ \alpha_q &= 2 - \cos q_x a - \cos q_y a + O(t) . \end{aligned} \qquad (10)$$

For $\vec{q}$ equal to $(2\pi(c-b)/b^2, 0)=\vec{q}_1$, a gap Δ is created in the phonon spectrum so that

$$\delta x_q \cong (\eta_q + \Delta\mu_q)/(\mathcal{J}\alpha_q + \Delta)\,. \tag{11}$$

A point needs to be emphasized. First of all because of the presence of the term $t_1 \sin[(2\pi/b)(i_x c)]$ in Eq. (4) the epitaxy exhibits an additional "pseudo-Bragg" peak at a wave vector $q_1 = 2\pi(c-b)/b^2$. Indeed, we can evaluate the structure factor S_q as follows:

$$S_q = \left\langle \sum_{l,j} \exp[i\vec{q}\cdot(\vec{r}_l - \vec{r}_j)] \right\rangle, \tag{12}$$

where the angular brackets denote a thermal average. Using Eq. (2), we can write

$$S_{q_x}(l,j) \equiv \exp[iq_x(x_l - x_j)] = \exp\{iq_x[c(l_x - j_x) + \delta x_l - \delta x_j]\}\exp(iq_x t\{\sin[2\pi(l_x c + \delta x_l)/b] - \sin[2\pi(j_x c + \delta x_j)b]\})\,. \tag{13}$$

Using an expansion analogous to equation (6), we get

$$S_{q_x}(l,j) \simeq \exp(iq_x[c(l_x - j_x) + \delta x_l - \delta x_j]) + \{J_0(tq_x) + 2iJ_1(tq_x)\sin[2\pi(l_x c + \delta x_l)/b] + \cdots\}$$
$$+ \{J_0(tq_x) - 2iJ_1(tq_x)\sin[2\pi(j_x c + \delta x_l)/b] + \cdots\}\,. \tag{14}$$

For $\vec{q}$ close to $\vec{q}_1$ only the second terms in the curly brackets are important, and we have, on substituting back into Eq. (12),

$$S_q \cong 4J_1^2(tq_x)J_0^2(tq_{1y}) \times \sum_{l,j} \exp[i(\vec{q}-\vec{q}_1)\cdot\vec{r}_{lj}] \times \exp[-(q_x - 2\pi/b)^2\langle(\delta x_{lj})^2\rangle]\,. \tag{15}$$

The dominant contribution to $\langle(\delta x_{lj})^2\rangle$ for $\vec{r}_{lj}$ large comes from the small q part of the phonon spectrum. Using Eq. (10), $\langle(\delta x_{lj}^2)\rangle$ can be evaluated and we have

$$\langle \delta x_{lj}^2 \rangle = (b^2/2\pi c)^2 \bar{\gamma}\, T \ln|\vec{r}_{lj}|\,,$$

where $\bar{\gamma} = \pi/\mathcal{J} + O(t)$. Thus

$$S_q \propto |\vec{q} - \vec{q}_1|^{\beta},$$

where

$$\beta = -2 + \bar{\gamma}(q_x - 2\pi/b)^2 T (b^2/2\pi c)^2\,. \tag{16}$$

Thus, as mentioned above, $S_{q_1} = \infty$ for $\bar{\gamma}T \leqslant 2$. We will see in a moment that this peak does disappear at a high enough temperature even though the above perturbation calculation is no longer valid.

III. THE HIGH-TEMPERATURE PHASE

Even in the commensurate case of $b=a$ for which the effect of the substrate is most important, the substrate potential becomes unimportant at a finite temperature. Indeed, for $b=a$, on making the substitution $x_i = i_x a + h_i$, the Hamiltonian (1) becomes the same as that studied in the roughening transition.[15] In that case, by doing a perturbation series in λ, it was found that the series converges at a high enough temperature whereas at low temperature it diverges term by term. Motivated by this analogy, we have carried out a perturbation calculation of λ for the case $b \neq a$. We have carried out this calculation explicitly to terms of the order of λ^4 and have placed some bounds for the general nth order term. The first nonvanishing term is of the order of λ^2 and is perhaps the most illustrative of the general behavior. The details of the calculation are very similar to our calculation on the roughening transition.[15] We tried to solve Eq. (3) perturbatively in λ. Defining a self-energy Σ by

$$\chi_{q\omega} = 1/(\alpha_q + \Sigma - i\omega/\Gamma)\,, \tag{17}$$

where χ is the response coefficient defined by

$$\chi_{q\omega} = \langle \Delta x_{q\omega}\rangle / \Delta\mu_{q\omega}\,, \tag{18}$$

we found in the static limit

$$\Sigma(q,0) \propto \lambda^2 \int d^2R\,(e^{i\vec{q}\cdot\vec{R}} - 1)R^{-\gamma T}\cos xq_{1x}\,. \tag{19}$$

Thus, if one examines the response near q_1, one finds that $\Sigma(q_1, 0)$ is divergent for low temperatures, consistent with our calculation in the previous section where a pseudo-Bragg peak occurs at $q = q_1$. Indeed, at $T=0$, Eq. (19) is just a delta function at $q = q_1$. For T larger than $T_2 = 2/\gamma$, however, the above expression for Σ is finite. Thus there is no Bragg peak at these temperatures. Note that for the roughening transition, the term $\cos q_{1x} x$ is replaced by one and the corresponding expression for Σ is finite for T larger than $T_1 = 4/\gamma$. That the perturbation series converges for $T > T_2$ rather than T_1 in the incommensurate case seems to be true to all orders of perturbation theory. The details of our calculation for the λ^4 term are reported in the Appendix, where we found that it is finite for $T>T_2$.

Actually, this problem is very similar to a problem concerning the localization of electrons in one dimension.[16] That problem can be mapped into a two-dimensional Coulomb gas in a random electric field in the x direction. An upper bound for the density autocorrelation function has been estimated, and it was shown that the perturbation series converges for $T<T_2'=3/\gamma$ independent of λ in the present language. The present problem can be mapped into a two-dimensional Coulomb gas with a constant imaginary electric field in the x direction,[17,18] and the arguments of Chui and Bray can be carried over directly, thus substantiating the claim that $T_2\neq T_1$. The above transition temperature is estimated in the limit of small λ. We expect T_2 to depend in general on λ, just like the two-dimensional Coulomb gas. To summarize, by doing perturbation calculations in terms of λ, we found that the response function diverges at $\vec{q}_1$ below T_2. Above T_2, however, it is finite.

IV. CONCLUSION

In this paper we propose that in the incommensurate phase in the limit of small λ for a high enough temperature ($T>T_2$), the substrate potential becomes suppressed. For T less than T_2, the structure factor S_q at $\vec{q}=\vec{q}_1=(2\pi(c-b)/b^2, 0)$ is infinite for $T<T_2$ but finite for $T>T_2$. The temperature T_2 is just half that of the roughening-transition temperature T_1. That something may be happening at the temperature T_2 is consistent with a recent work of Luther, Timonen, and Pokrovsky,[19] but our physical picture is quite different. Luther *et al.* mapped the present problem into a fermion problem with the Fermi surface shifted, and extracted their results from that picture. Chui and Bray[17] have also mapped the present problem into one with an additional electron-electron interaction. That problem is not inconsistent with that with a shifted Fermi surface, but there are differences. Chui and Bray have associated the Bragg peak mentioned above with the occurrence of the $4k_F$ correlation function in the one-dimensional electron-gas problem (which could be interpreted as due to a shifted Fermi surface).

It is tempting to speculate what the phase diagram of our model will be. This is illustrated in Fig. 1. At $T=0$ there is a CIT occurring at point C. For the commensurate case ($b=a$) there is a "floating solid transition"[20] analogous to roughening transition occurring at a finite temperature A. The two incommensurate regions that we discussed here are separated by the line BD.

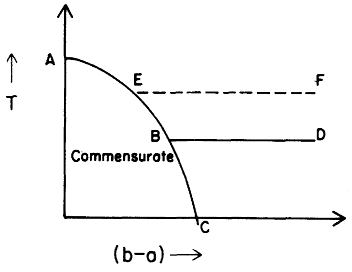

FIG. 1. Possible schematic phase diagram.

If the substrate potential is suppressed then the lattice constants may change discontinuously from the commensurate to the incommensurate phase. Thus it is possible that AB is a first-order phase-transition boundary. EF is the two-dimensional melting transition boundary discussed recently by Halperin and Nelson.[20] The position of EF depends on the density of dislocations whereas that of BD depends on λ, so *a priori* we do not know the relative position between these two. We have assumed that EF occurs above BD so that the effect of dislocations is simply to renormalize $\mathcal{J}$.[20] If EF occurs below BD, then the situation becomes more complicated and a more careful analysis is necessary.

In real physical systems it may not be possible to reach A because of the formation of two or more layers. Also the interepitaxial atomic potential is quite anharmonic. Such effects have not been taken into account here. Indeed we think that they can account for some of the inconsistencies between theory and experiment in the CIT.

ACKNOWLEDGMENT

This work was supported in part by the National Science Foundation under Grant No. DMR-7918697.

APPENDIX

In this appendix the calculation of the fourth-order term is carried out. As we mentioned, the present problem can be mapped into that of a two-dimensional Coulomb gas in an imaginary electric field. From that we know that the quantity of interest is proportional to $\int \exp(i\vec{q}_1\cdot\vec{R})\, I_0(R)d^2R$, where $I_0(R)$ is given by

$$I_0(R)=\int_0^R \prod_{i=2}^{4} d^2r_i\left(\frac{r_{12}r_{34}}{r_{13}r_{14}r_{23}r_{24}}\right)^{\gamma} \times\exp[iq_{1x}(x_1+x_2-x_3-x_4)]\,. \qquad \text{(A1)}$$

Choosing particle 1 to be the origin and expanding the exponentials in terms of Bessel functions, we get

$$I_0=R^2\int \prod_{i=2}^{4} d^2r_i\left(\frac{r_2}{r_3r_4}\right)\left(\frac{|r_3-r_4|}{|r_2-r_3|\,|r_2-r_4|}\right)^{\gamma}$$
$$\times\sum_{m_2,m_3,m_4}\prod_{j=2}^{4}J_{m_j}(q_{1x}|r_i|)$$
$$\times i^{m_2+m_3+m_4}\exp\left(i\sum_{j=2}^{4}m_j\theta_j\right). \qquad \text{(A2)}$$

$|\vec{R}-\vec{R}'|^{-\gamma}$ can be expanded in terms of the Gegenbauer polynomials C_n^{γ} as

$$|\vec{r}-\vec{r}'|^{-\gamma}=\sum_n r_>^{-\gamma-n}r_<^{n}C_n^{\gamma}(\cos(\theta-\theta')). \qquad \text{(A3)}$$

We now evaluate I_0 using spherical coordinates. The radial part of the integral can be split up into different regions as

$$\int=\int_{R>r_2>r_3>r_4}+\int_{R>r_3>r_2>r_4}+\int_{R>r_2>r_4>r_3}+\int_{R>r_3>r_4>r_2}+\int_{R>r_4>r_2>r_3}+\int_{R>r_4>r_3>r_2}$$
$$=\sum_{\substack{n_1,n_2,n_3\\ \epsilon_1,\epsilon_2,\epsilon_3=\pm}}\sum_{i=1}B_i\begin{bmatrix}\epsilon_1 & \epsilon_2 & \epsilon_3\\ n_1 & n_2 & n_3\end{bmatrix}I_a(n_1n_2n_3)[q_{1x}(2/\pi)^{1/2}]^3, \qquad \text{(A4)}$$

where, for example,

$$B_1\begin{bmatrix}+ & + & +\\ n_1 & n_2 & n_3\end{bmatrix}=\int_{R>r_2>r_3>r_4}\prod dr_i\cos(q_{1x}r_2-\pi/4)\cos(q_1r_3-\pi/4)\cos(q_1r_4-\pi/4)$$
$$\times r_3^{-n_1+n_2+1/2}r_4^{-\gamma+n_1+n_3+1/2}r_2^{-n_2-\gamma-n_3+1/2}. \qquad \text{(A5)}$$

The terms with "$-$" can be obtained by replacing the corresponding cos by sin. Since we are only interested in whether the integral diverges, the dominant contribution to the integral comes only when $|r_i|$ is large. In (A4) and (A5) we have used the asymptotic expansion of the Bessel functions, viz.,

$$J_m(x)\to(2/\pi x)^{1/2}\cos(x-\tfrac{1}{4}m\pi-\tfrac{1}{4}\pi). \qquad \text{(A6)}$$

I_a in Eq. (A4) comes from the angular integration. The following series representation of the Gegenbauer polynomials have been used:

$$C_n^{\alpha}(\cos\theta)=\sum_{m=0}E\begin{bmatrix}\alpha & & r\\ & m & \end{bmatrix}\cos(r-2m)\theta, \qquad \text{(A7)}$$
$$E\begin{bmatrix}\alpha & & r\\ & m & \end{bmatrix}=\frac{\Gamma(\alpha+m)\Gamma(\alpha+r-m)}{m!\,(n-m)!\,[\Gamma(\alpha)]^2}.$$

I_a is given by

$$I_a=\sum_{m_2,m_3,m_4}\prod_i\tfrac{1}{2}[1+\epsilon_i(-)^{m_i}]I_A, \qquad \text{(A8)}$$
$$I_A=\sum_{p_i=0}^{n_i}E\begin{bmatrix}-\gamma/2 & & n_1\\ & p_1 & \end{bmatrix}E\begin{bmatrix}\gamma/2 & & n_2\\ & p_2 & \end{bmatrix}E\begin{bmatrix}\gamma/2 & & n_3\\ & p_3 & \end{bmatrix}i^{m_2+m_3+m_4}\,\delta(m_2-|r_2-2p_2+r_3-2p_3|)$$
$$\times[\delta(m_4-|r_3-2p_3-r_1+2p_1|)\delta(m_3-|n_1-2p_1+r_2-2p_2|)$$
$$+\delta(m_4-|r_1-2p_1+r_3-2p_3|)\delta(m_3-|2p_1-r_1+r_2-2p_2|)+\delta(m_4-|r_3-2p_3-r_1+2p_1|)]$$
$$\times[\delta(m_4-|r_3-2p_3-r_1+2p_1|)\delta(m_3-|-r_1+2p_1+r_2-2p_2|)$$
$$+\delta(m_4-|r_1-2p_1+r_3-2p_3|)\delta(m_3-|r_1-2p_1+r_2-2p_2|)]. \qquad \text{(A9)}$$

The delta function comes from the integration over all the $\cos\theta$ factors.

On explicitly taking care of the delta functions, I_a can be simplified as

$$I_a=\tfrac{1}{2}C_{n_1}^{-\gamma/2}(1)C_{n_2}^{\gamma/2}(1)C_{n_3}^{\gamma/2}(1)[1+\epsilon_2(-)^{n_2+n_3}][1+\epsilon_4(-)^{n_3+n_1}][1+\epsilon_3(-)^{n_1+n_2}]. \qquad \text{(A10)}$$

Putting (A10) back into (A4) we get

$$I_0 = \sum_i F_i(r_1, r_2, r_3)[q_1(2/\pi)^{1/2}]^3 C_{r_1}^{-\gamma/2}(1) C_{r_2}^{\gamma/2}(1) C_{r_3}^{\gamma/2}(1)\,, \tag{A11}$$

where, for example,

$$\begin{aligned} F_1(n_1, n_2, n_3) = i^{2(n_1+n_2+n_3)} \int_{R>r_2>r_3>r_4} & \prod dr_i \cos[q_{1x} r_2 - \tfrac{1}{4}\pi - \tfrac{1}{2}(n_2+n_3)\pi] \\ & \times \cos[q r_3 - \tfrac{1}{4}\pi - \tfrac{1}{2}(n_2+n_1)\pi] \cos[q r_4 - \tfrac{1}{4}\pi - \tfrac{1}{2}(n_3+n_1)\pi] \\ & \times r_3^{-n_1+n_2+1/2} r_4^{-\gamma+n_1+n_3+1/2} r_2^{-n_2-\gamma-n_3+1/2}\,. \end{aligned} \tag{A12}$$

The subscript of F indicates the different region of integration as is illustrated in equation (A4). Because of the cos factors in (A12), the integrals F_i converge as $R \to \infty$ whenever the integrand is a decreasing function of R. If the cos factors are absent, the phase-space factors $\Pi_i\, dr_i$ have also to be counted in order to look for a divergence. This corresponds exactly to the difference between T_2 and T_1 that we encountered in the second-order calculation.

One might wonder whether, because of the infinite summation in (A11), that even though F_i is well defined, I_0 can "still" be divergent. To clarify this point we assume r_i to be very large and use the asymptotic expansions for the Γ functions to obtain

$$C_{n_1}^{\gamma}(1) \to n_1^{2\gamma-1} \tag{A13}$$

and

$$I_0 \cong \sum_i F_i(n_1, n_2, n_3) n_1^{-2\gamma-1} n_2^{2\gamma-1} n_3^{2\gamma-1}\,. \tag{A14}$$

The integrals F_i can be estimated using the incomplete gamma function and its asymptotic expansion, viz.,

$$\int^x y^{\mu-1} \cos\beta y\, dy = \tfrac{1}{2}[(i\beta)^{-\mu}\gamma(\mu, i\beta x) + (i\beta)^{-\mu}\gamma(\mu, -i\beta x)]\,, \tag{A15}$$

with a similar expression for the sin and

$$\lim_{x\to\infty} \gamma(\alpha, x) = \Gamma(\alpha) - x^{\alpha-1}e^{-x}\,. \tag{A16}$$

On substituting (A16) and (A15) into (A12) we get, as far as the n dependence is concerned,

$$\begin{aligned} F_1(n_1, n_2, n_3) \propto \Gamma(-n_1+n_2-\tfrac{1}{2})\Gamma(-\gamma+n_1+n_3-\tfrac{1}{2}) \\ \times \Gamma(-n_2-\gamma-n_3-\tfrac{1}{2})\,. \end{aligned}$$

In the limit in which all three n_i's are large, say, $n_1 = \alpha_1 N$, $n_2 = \alpha_2 N$, $n_3 = \alpha_3 N$, where $N \to \infty$, we found that

$$I_0 \propto \sum_{\alpha_1, \alpha_2, \alpha_3} \sum_N N^{-2\gamma-3} \propto \sum_{\alpha_1 \alpha_2 \alpha_3} N^{-6}\,.$$

Since the different choices of α_1, α_2, α_3 will provide a phase-space factor proportional only to N^3, the resulting series for I_0 is still convergent, however, thus justifying our claim.

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