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Eigenmodes of the coupled two-dimensional Wigner-crystal–liquid-surface system and instability of a charged liquid surface

André-Marie Tremblay and Vinay Ambegaokar
Laboratory of Atomic and Solid State Physics
and Materials Science Center, Cornell University, Ithaca, New York 14853
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The coupled phonon–surface-wave modes of a two-dimensional Wigner crystal resting on the surface of a liquid are studied. It is shown that, in experimentally accessible situations, the threshold for the formation of the macroscopic "dimple crystal" predicted by Gor'kov and Chernikova is negligibly influenced by the presence of the Wigner crystal, and that the coupled phonon–short-wavelength-surface waves studied recently by Fisher, Halperin, and Platzman, are negligibly influenced by the soft mode which leads to the "dimple crystal".

I. INTRODUCTION

Recent experiments[1] supported by a quantitative theoretical interpretation[2] have demonstrated that electrons trapped above the surface of helium can form a two-dimensional Wigner crystal. In the theory, the detected modes are convincingly interpreted as coupled umklapp phonon-ripplon modes, i.e., modes which couple electron-lattice vibrations and helium surface waves with wave numbers differing by vectors of the reciprocal lattice.

Recently, Wanner and Leiderer[3] also observed a "macroscopic" Wigner crystal, or "dimple crystal", in a system of ions at the interface of a ^{3}He-^{4}He mixture. The results of this experiment had been predicted by Gor'kov and Chernikova[4] on the basis of a coupled plasmon–long-wavelength-ripplon soft mode. Wanner and Leiderer have suggested that in their experiment, the dimple crystal may coexist with the Wigner crystal.

In this paper we study the coupled electron-crystal–surface-wave system including both the long-wavelength and the umklapp ripplon-phonon modes. We show that the effect of the "self-induced" field, which is very important for the Gor'kov-Chernikova instability,[4] can safely be neglected in the study of umklapp phonon-ripplon modes, as has been assumed in Ref. 2. We also show that, for experimentally realizable systems, the long-wavelength modes, and in particular the Gor'kov-Chernikova[4] soft mode, are decoupled from the umklapp phonon-ripplon modes[2] of the Wigner crystal.

II. EQUATIONS OF MOTION

Assuming that the wavelengths of interest are much smaller than the helium depth,[5] the equation of motion for the Fourier component of the displacement ξ of the helium surface is[6]

$$\frac{-\omega^2\rho\xi_{\vec{q}}}{q}+\sigma q^2\xi_{\vec{q}}+\rho g\,\xi_{\vec{q}} = \int d^2r\, e^{-i\vec{q}\cdot\vec{r}}[eE_\perp(\vec{r})\,n(\vec{r})]+U_{\vec{q}}\,n_{\vec{q}}\ , \tag{1}$$

where ρ is the density of helium, σ is its surface tension, n is the electron density, and g is the gravitational acceleration. The last term in Eq. (1) represents the pressure due to the monopole–induced-dipole force between the electrons and the helium atoms.[7] (It reduces to the usual image charge effect when the helium surface is flat.) The first term on the right-hand side of Eq. (1) is the pressure due to the electric field perpendicular to the helium surface. To zeroth order,

$$eE_\perp^0 = 2\pi e^2 n_s + e\mathcal{E}_\perp\ , \tag{2}$$

where $\mathcal{E}_\perp$ is an "external clamping field" and $2\pi en_s$ is the field due to the neutralizing positive charge of density n_s on the plate at the bottom of the helium container.

The integral in Eq. (1) is over the two-dimensional helium surface, and the Fourier transforms are de-

fined as follows:

$$n_{\vec{q}} \equiv \int d^2r\, e^{-i\vec{q}\cdot\vec{r}} n(\vec{r}) \ , \tag{3}$$

$$n(\vec{r}) \equiv \int \frac{d^2q'}{(2\pi)^2} e^{i\vec{q}'\cdot\vec{r}} n_{\vec{q}'} \equiv \frac{n_s}{N} \sum_{\vec{q},\vec{G}} e^{i(\vec{q}+\vec{G})\cdot\vec{r}} n_{\vec{q}+\vec{G}} \ . \tag{4}$$

In the last expression, $\vec{q}$ is restricted to the first Brillouin zone and $\vec{G}$ runs over all reciprocal-lattice vectors. N is the number of unit cells in the crystal. When, in what follows, sums over lattice sites $\vec{R}_i$ occur, we shall use the following pair of transforms:

$$u_{\vec{q}} = \frac{1}{N} \sum_{R_i} e^{-i\vec{q}\cdot\vec{R}_i} u(\vec{R}_i) \ , \tag{5}$$

$$u(\vec{R}_i) = \sum_{\vec{q}} e^{i\vec{q}\cdot\vec{R}_i} u_{\vec{q}} \ . \tag{6}$$

Here $\vec{q}$ is confined to the first Brillouin zone.

Equation (1) has a static solution, $\xi_G^0 \neq 0$ reflecting the fact that each electron depresses the helium around itself.[8]

Now we allow for the electrons to move around their equilibrium positions $\vec{R}_i$ by a small amount $\vec{u}(\vec{R}_i)$ in the horizontal plane. The displacement of a given electron perpendicular to the plane will equal the displacement ξ of the helium surface underneath it because the vertical motion of the electron with respect to the helium surface is quantized[9] and the binding energy of the lowest state is much larger then the energies involved in the modes we shall be studying.

The equation of motion for the electron displacements in the horizontal plane is[2]

$$-\omega^2 u_q^{\alpha} = -\omega_{\alpha\beta}^2 u_q^{\beta} + \frac{in_s}{mN} \sum_G (eE_\perp^0 + U_{q+G})(q+G)^{\alpha} n_{q+G}^0 \xi_{q+G} - \left(\frac{n_s}{m\sigma}\right) \sum_G (eE_\perp^0 + U_G)^2 \frac{G^{\alpha}G^{\beta}}{G^2} n_G^{0^2} u_q^{\beta} \ , \tag{7}$$

where m is the electron mass, α and β label the directions, and n_q^0 is the "form factor"[2]

$$n_G^0 = e^{-G^2\langle u_z^2\rangle/4} \ . \tag{8}$$

When G is the shortest reciprocal-lattice vector,[2] $n_G^0 \sim 0.5$. The first term on the right-hand side of Eq. (7) describes the electron system on a flat helium surface, while the last two terms represent phonon-ripplon interactions. The last term in Eq. (7) is due to the restoring force from the static deformation of the helium surface,

$$\xi_G^0 = (eE_\perp^0 + U_G) N n_G^0/\sigma G^2 \quad (\xi_{G=0}^0 \equiv 0) \ .$$

We now expand the right-hand side of Eq. (1) to first order in the electron displacements. The electron displacement along the horizontal plane $\vec{u}(\vec{R}_i)$ will produce changes in the density $n(\vec{r})$ while the displacement perpendicular to the helium plane will induce a field $eE_\perp^1$. It is this last term which we call the "self-induced field". Taking all these effects into account, the equation of motion for $\xi_{\vec{q}}$ becomes

$$(-\omega^2 + \Omega_q'^2)\xi_q = -N(eE_\perp^0 + U_q)\, i\vec{q}\cdot\vec{u}_{\vec{q}} n_q^0 q/\rho + 2\pi e^2 n_s^2 q/\rho \sum_{G_1 G_2}{}' (|-\vec{q}+\vec{G}_1| - |\vec{G}_1|)\, n_{G_1}^0 n_{-G_1-G_2}^0 \xi_{q+G_2} \ , \tag{9}$$

where

$$\Omega_q'^2 = (\sigma/\rho)q^3 + gq \tag{10}$$

and $\vec{q}$ in $u_{\vec{q}}$ must be folded back to the first Brillouin zone. The last term of Eq. (9) represents the effect of the "self-field". In Ref. 2 this term was neglected.

III. EIGENFREQUENCIES

In the long-wavelength limit, the dynamical matrix $\omega_{\alpha\beta}$ may be diagonalized (with an accuracy of order q/G) by taking one polarization vector along the direction of q and the other perpendicular to it.[10] To leading order in q/G these will remain the directions of the eigenvectors for the full system of coupled Eqs. (7) and (9).

Equations (7) and (9) have several solutions that do not involve any electron motion[2] ($\vec{u}=0$). Suppose there exist k reciprocal-lattice vectors with magnitude $G(G \neq 0)$. There are $k-2$ solutions to our system of equations which have eigenfrequency $\omega = \Omega_G'$ (to leading order in q/G) and eigenvector determined from the equations,

$$\begin{aligned} &\xi_{q+G'} = 0, \quad \text{for } G' \neq G \ , \\ &\sum_{\langle G\rangle} \xi_{q+G}(q+G)^{\lambda}(eE_\perp^0 + U_{q+G}) n_{q+G}^0 = 0 \ . \end{aligned} \tag{11}$$

where λ represents the longitudinal (l) or transverse (t) direction and $\sum_{\langle G\rangle}$ means sum over all possible *directions* of the reciprocal-lattice vector of magnitude G.

There will be two more eigenfrequencies around $\omega \sim \Omega_G$; one will correspond to coupled longitudinal phonon-ripplon coordinates and the other to coupled transverse phonon-ripplon coordinates. For $\omega \sim \Omega_q$ there is only one mode and it involves longitudinal phonons and ripplons. This will be the only mode for which the self-field contribution will be important.

We proceed to find these coupled-mode eigenfrequencies. Solving Eq. (7) for $\vec{u}$ and substituting in Eq. (9) we find,

$$\xi_{q+G}=\frac{V^0_{q+G}}{\nu^2_\lambda}n^0_{q+G}\frac{(q+G)^\lambda}{|q+G|^2}\frac{\Omega^2_{q+G}}{-\omega^2+\Omega'^2_{q+G}}\sum_{G'}(q+G')^\lambda\xi_{q+G'}n^0_{q+G'}V^0_{q+G'}$$
$$+\frac{2\pi e^2n_s^2|q+G|}{-\omega^2+\Omega'^2_{q+G}}\frac{1}{\rho}\sum_{G'}\left[qn^0_0n^0_{-G'}-\sum_{G_1\neq 0}\frac{\vec{q}\cdot\vec{G}_1}{G_1}n^0_{G_1}n^0_{-G'-G_1}\right]\xi_{q+G'}\ . \tag{12}$$

The terms in the last sum of Eq. (12) have been expanded to lowest order in q/G. Following Fisher *et al.*[2] we defined in Eq. (12)

$$V^{0^2}_q=(eE^0_\perp+U_q)^2\frac{n_s}{m\sigma}\ , \tag{13}$$

and

$$\omega^2_d=\sum_G\frac{G^{\lambda^2}}{G^2}V^{0^2}_G n^{0^2}_G\ , \tag{14}$$

$$\Omega^2_q=\Omega'^2_q-gq\ , \tag{15}$$

$$\nu^2_\lambda=-\omega^2+\omega^2_\lambda(q)+\omega^2_d\ , \tag{16}$$

$$\omega^2_{\lambda=l}(q)=\omega^2_l(q+G)=\frac{2\pi e^2n_sq}{m}\ , \tag{17}$$

$$\omega^2_{\lambda=t}(q)=\omega^2_t(q+G)\equiv s^2q^2\ . \tag{18}$$

The transverse velocity of sound s is given in Ref. 10.

In the Appendix, we derive sufficient conditions for the self-field terms in Eq. (12) to be smaller than the terms proportional to V^0_{q+G}. In most instances, the only non-negligible effect of the self-field is to renormalize the lowest bare ripplon frequency Ω'_q:

$$\tilde{\Omega}^2_q=\Omega'^2_q-2\pi e^2n_s^2q^2/\rho\ . \tag{19}$$

From Eq. (12) we thus find that, to leading order in (q/G), the dispersion relation is

$$\omega^2-\omega^2_\lambda(q)-\sum_{G\neq 0}\frac{G^{\lambda^2}}{G^2}V^2_G\frac{\omega^2}{\omega^2-\Omega^2_G}$$
$$-\frac{q^{\lambda^2}}{q^2}V^2_q\frac{\Omega^2_q}{\omega^2-\tilde{\Omega}^2_q}=0\ , \tag{20}$$

where we defined $V_G\equiv V^0_Gn^0_G$. The last term in Eq. (20), representing the effect of the low-frequency ripplons, is not present in Ref. 2. The Gor'kov-Chernikova limit (electron liquid) can be obtained from the fact that in that limit, $n^0_q\sim 1$ and $n^0_G=0$. Note that the last term of Eq. (20) vanishes when $\lambda=t$. We can also check that our choice of directions of the eigenvectors is self-consistent.

To solve Eq. (20) it is useful to note that

$$\frac{\tilde{\Omega}_q{}^2}{\omega^2_l(q)}<\frac{\Omega'^2_q}{\omega^2_l(q)}=\frac{gq+\sigma q^3/\rho}{\omega^2_l(q)}<\frac{2gq_c}{\omega^2_l(q_c)}\ . \tag{21}$$

To verify the last inequality, use the fact that the ratios are monotonically increasing functions of q and that the largest q of interest, q_c, is at the same time the soft-mode wave vector and 2π divided by the capillary length. Thus,

$$\frac{\tilde{\Omega}_q{}^2}{\omega^2_l(q)}<\frac{2gq_c}{\omega^2_l(q_c)}=\frac{2mg}{2\pi e^2n_s}<<1\ . \tag{22}$$

This ratio is small for any reasonable density n_s since it is the ratio between a gravitational and an electrical force.

Ignoring the effect of the lattice, i.e., the sum over G in Eq. (20), and using Eq. (22), we find the low-frequency mode of Gor'kov and Chernikova[4]

$$\omega^2=\tilde{\Omega}_q{}^2-\frac{V^2_q\Omega^2_q}{\omega^2_l(q)}\ . \tag{23}$$

The corrections to this result arising from the lattice are of order

$$\frac{3V^2_G[\tilde{\Omega}_q{}^2-V^2_q\Omega^2_q/\omega^2_l(q)]}{\omega^2_l(q)\Omega^2_G}<\frac{3V^2_G\Omega^2_{q_c}}{\omega^2_l(q_c)\Omega^2_G}$$
$$\sim\frac{V^2_{q_c}\Omega^2_{q_c}}{\omega^2_l(q_c)\Omega^2_G}\lesssim\frac{\tilde{\Omega}_{q_c}{}^2}{\Omega^2_G}$$
$$<\frac{\Omega'^2_{q_c}}{\Omega^2_G}\sim\left[\frac{q_c}{G}\right]^3<<1\ , \tag{24}$$

where we used the fact that the first ratio in Eq. (24) is a monotonically increasing function of q (we are interested in $q<q_c$) and that $\omega^2>0$ in Eq. (23). The Gor'kov-Chernikova mode[4] is thus negligibly influenced by the presence of the lattice.

Similarly, we find that the umklapp phonon-ripplon modes[2] are negligibly influenced by the Gor'kov-Chernikova mode. For example, if G_1 is the shortest reciprocal-lattice vector, then the correction to

$$\omega^2\sim\Omega^2_{G_1}[1+3V^2_{G_1}/\omega^2_l(q)]^{-1} \tag{25}$$

is of order $V^2_q\Omega^2_q/\omega^2_l(q)\Omega^2_G$ which by Eq. (24) can be neglected for the whole range of values of q of interest.

Finally, for $\omega\sim\omega_l(q)<<\Omega_{G_1}$, we find that the root

$$\omega^2=\left[1+\sum_{G\neq 0}\frac{G^{\lambda^2}}{G^2}\frac{V^2_G}{\Omega^2_G}\right]^{-1}\omega^2_l(q)\equiv\frac{m}{m^*}\omega^2_l(q) \tag{26}$$

is negligibly modified by the presence of the low-frequency ripplon mode because

$$\frac{m^*}{m}\frac{V^2_q\Omega^2_q}{\omega^4_l(q)}<\frac{m^*}{m}\frac{\tilde{\Omega}_q{}^2}{\omega^2_l(q)}<\frac{m^*}{m}\frac{\Omega'^2_q}{\omega^2_l(q)}<<1\ , \tag{27}$$

where the first inequality is a consequence of $\omega^2 > 0$ in Eq. (23), and the last inequalities a consequence of Eqs. (21) and (22).

Note that since the unperturbed frequencies Ω_q' and $\omega_l(q)$ both vanish as $q \to 0$, it is not obvious *a priori* that their interaction can be neglected when $q \to 0$.

IV. DISCUSSION OF OTHER SYSTEMS AND CONCLUDING REMARKS

Our results depend mainly on the fact that the ratio of the soft-mode wave vector q_c and a reciprocal-lattice vector G is small. More interesting situations might occur when the ratio becomes of order unity, as, for example, is the ratio of the Fermi wave vector and a reciprocal-lattice vector in the Peierls instability problem. We looked for systems where the case $q_c/G \sim 1$ could be achieved: electrons on He, H, Ne, and ions on the interface of ^{3}He-^{4}He mixtures. Systems for which the surface tension can be made very small may lead to large ratios of q_c/G. Small surface tensions may occur at phase transitions. Figure 1 shows the phase diagram for ions on ^{3}He-^{4}He mixtures for which the surface tension vanishes at

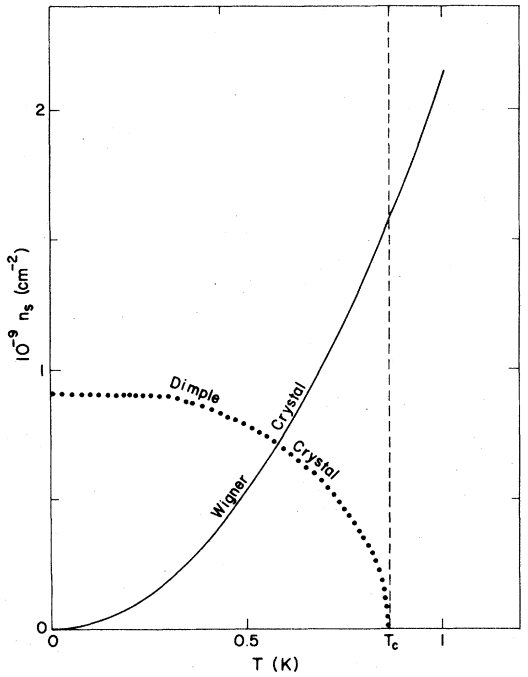

FIG. 1. Phase diagram for ions on the interface of phase separated ^{3}He-^{4}He mixtures. The dotted line is the Gor'kov-Chernikova instability threshold line. In the neighborhood above that line, a "dimple crystal" can exist. Above the solid line, a classical Wigner crystal of ions can exist. At $T = T_c$ the surface tension vanishes. The line $(q_c/G)^2 = 1$ is, on this scale, indistinguishable from the vertical line at $T = T_c$.

$T_c = 0.867$ K. The dotted line is the Gor'kov-Chernikova instability threshold line[11] $n_s = [(\rho g \sigma)^{1/2}/2\pi e^2]^{1/2}$. In the neighborhood above that line, a "dimple crystal" can exist. Above the solid line,[1] $n_s > (\Gamma k_B T/\pi^{1/2} e^2)^2$ ($\Gamma = 137$), a classical Wigner crystal of ions can exist. (The straight line $\pi \hbar^2 n_s/m_i = k_B T$, which indicates where quantum-mechanical effects may become important, has not been plotted here because it is above the classical melting curve for the range of temperatures of interest.) The ratio q_c/G_1 diverges at $T = T_c$ where the interface between ^{3}He and ^{4}He disappears. The line $(q_c/G_1)^2 = (\rho g/\sigma)/[8\pi^2 n_s/(3)^{1/2}] = 1$ intersects the Wigner-crystal melting curve at $T_c - T \sim 10^{-4}$ and goes below $n_s = 10^{-7}$ around $T_c - T \sim 10^{-3}$. On this scale, it can thus not be resolved from the vertical dashed line at $T = T_c$. We thus find that the condition $q_c/G << 1$ is always satisfied except in a very narrow region along the part of the vertical dashed line which is above the solid line. In that region, the Wigner crystal and the dimple crystal would interact strongly with each other. It is not obvious what order would then occur since, in addition to the strong interaction between the crystals, the following difficulties may also arise: (a) The dimple crystal might not be stable and have a lattice vector q_c when it is very far away from the instability threshold. (b) The Wigner crystal might not remain stable because the interface thickness diverges at $T = T_c$.

Note that the "dimple crystal" can exist at lower densities if a clamping field $\mathcal{E}_\perp$ is used. If the direction of $\mathcal{E}_\perp$ is changed, the instability might occur at slightly higher densities. The increase however is limited by the presence of the self-field terms which cannot be "turned off".

Qualitatively similar results are obtained for the case of electrons on *liquid* He, H, and Ne. Note that in the last two cases there may be important quantum-mechanical effects in the electronic system before the Wigner crystal forms.

It is thus likely that in physical systems of interest, the condition $q_c/G << 1$ will be satisfied, in which case, the main results of this paper should apply.

In summary, we have shown that the self-induced field has a negligible effect on the umklapp phonon-ripplon modes[2] and that the Gor'kov-Chernikova[4] soft mode is negligibly influenced by the presence of the Wigner lattice. That conclusion depends mainly on the fact that the ratio of the soft-mode wave vector q_c and a reciprocal-lattice vector G is small.

Note added in proof. P. Leiderer (private communication) has pointed out that another factor which makes it impossible to study a Wigner crystal of ions at the interface of ^{3}He-^{4}He mixtures very close to $T_c = 0.867$ K is that the trapping time of ions becomes vanishingly small in the limit $T \to T_c$. [M. Wanner, P. Leiderer, and T. Reverchon, Phys. Lett. A 68, 226 (1978).]

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APPENDIX: COMPARISON OF SELF-FIELD TERMS AND TERMS PROPORTIONAL TO V^0_{q+G} IN EQ. (12)

The terms containing V^0_{q+G} (type-I terms) in Eq. (12) are those that have been considered by Fisher *et al.*[2] A term by term comparison of type-I terms and terms arising from the self-field (type-II terms) shows that type-II terms may become larger than type-I terms only for special directions of G and G'. However, in Fisher's solution, the values of ξ in these directions are smaller than the values of ξ in most other directions by a factor q/G.

A more quantitative comparison proceeds as follows. Consider the Fisher *et al.*[2] solution

$$\xi^0_{q+G}=\frac{V^0_{q+G}}{\nu^2_\lambda}n^0_{q+G}\frac{(q+G)^\lambda}{|q+G|^2}\frac{\Omega^2_{q+G}}{-\omega^2+\Omega'^2_{q+G}}A$$

$$\sim B(|G|)(G)^\lambda , \tag{A1}$$

where A is a constant and B depends on the magnitude but not the direction of G. We can check that the terms of type II in Eq. (12) are much less than those of type I (in a perturbative sense) by substituting Eq. (A1) into Eq. (12) and comparing the $\sum_{\langle G'\rangle}$ from type-II terms with the $\sum_{\langle G'\rangle}$ from type-I terms. The evaluation of type-II terms is simplified by the following rough estimate, based in part on the fact that n^0_G is an exponentially decreasing function of G,

$$\sum_{G_1\neq 0}\frac{-\vec{q}\cdot\vec{G}_1}{G_1}n^0_{G_1}n^0_{-G-G_1}\sim\frac{\vec{q}\cdot\vec{G}}{G}n^0_{-G}n^0_0 . \tag{A2}$$

Note that the self-field term for the case $\lambda=t$ vanishes identically if it is estimated using Eqs. (A1) and (A2). The comparison that follows is thus restricted to the case $\lambda=l$. We finally find that when $G'\neq 0$, type-II terms are smaller than type-I terms by at least a factor,

$$\frac{\nu_l^2}{V_q^2}\frac{2\pi e^2 n_s^2}{\sigma G'}=\frac{\nu_l^2}{V_q^2}\left[\frac{2\pi e^2 n_s^2 q_c^2}{\rho\Omega^2_{q_c}}\right]\frac{q_c}{G'}\sim 10^{-3} , \tag{A3}$$

where, from Eq. (23), the quantity in brackets is of order unity at the largest achievable densities. The numerical estimate in Eq. (A3) applies to the Grimes and Adams experiment.[1] In that experiment, $\nu_l^2/V_q^2<10^2$ when $q\leq q_c$.

It is more delicate to compare the contribution from the $G'=0$ terms. In fact when $G=0$, type-II terms with $G'=0$ renormalize the frequency Ω'_q by an amount $\tilde{\Omega}_q-\Omega'_q$ [see Eq. (19)] which, when $e\mathcal{E}_\perp+U_q<<2\pi e^2 n_s$, is about equal to the renormalization from type-I terms.

When $G\neq 0$ we find that, for certain directions of G, the $G'=0$ contributions from type-II terms may again be larger than those from type I. If we do not neglect these terms, we find that the resulting corrections to the dispersion relation could be important only when $\omega\sim\tilde{\Omega}_q$. But if we substitute the Gor'kov-Chernikova result Eq. (23), we find, again by comparing various $\sum_{\langle G'\rangle}$, that these corrections are of order

$$\frac{2\pi e^2 n_s^2}{\sigma G}\left[\frac{q}{G}\right]\left[\frac{\omega_l^2(q)}{\nu_l^2}\right] ,$$

which is even smaller than the factor in Eq. (A3).

Finally, note that one could apply an external field such that $V_q\sim 0$, as can be checked from Eqs. (2) and (13). As Eqs. (12) and (A3) indicate, in such a case the self-field terms would dominate (although the above perturbation technique cannot strictly be used to show that). All of this is inconsequential however since (a) we expect the electrons to evaporate when $V_q=0$; (b) it can be proved that, even when $V_q=0$, the effect of the self-field terms on the modes at $\omega\sim\Omega_G$ is negligible.

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