Ginsburg-Landau theory of supersolid

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We develop a simple Ginsburg-Landau theory to study all the possible phases and phase transitions in $^4$He, analyze the condition for the existence of the supersolid (SS) and map out its global phase diagram from a unified framework. If the condition favors the existence of the SS, we use the GL theory to address several experimental facts and also make some predictions that are amenable to experimental tests. A key prediction is that the X-ray scattering intensity from the SS ought to have an additional modulation over that of the NS. The modulation amplitude is proportional to the Non-Classical Rotational-Inertial (NCRI) observed in the torsional oscillator experiments.

1. Introduction: A solid can not flow. While a superfluid can flow without any resistance. A supersolid (SS) is a new state of matter which has both the solid and superfluid order. The possibility of a supersolid phase in $^4$He due to very large zero point quantum fluctuations was theoretically speculated in 1970 [1–4]. Over the last 35 years, a number of experiments have been designed to search for the supersolid state without success. However, recently, by using torsional oscillator measurement, a PSU group lead by Chan observed a marked $1\%$ increase in structure factor [5]. The second is the phenomenological approach [6]. The authors suggested that the supersolid state is responsible for the NCRI. The PSU experiments rekindled extensive theoretical interests [6–8] in the still controversial supersolid phase of $^4$He. There are two kinds of complementary theoretical approaches. The first is the microscopic numerical simulation [6]. The second is the phenomenological approach [7,8]. In this paper, by constructing a Ginsburg Landau (GL) theory, we will address the following two questions: (1) What is the condition for the existence of the SS state? (2) If the SS exists, what are the properties of the supersolid to be tested by possible new experiments. We develop a simple GL theory to map out the $^4$He phase diagram and study all the phases and phase transitions in a unified framework. If the repulsive coupling in Eqn.3 in the GL theory is sufficiently small, the SS phase becomes stable at low enough temperature. The resulting solid at high pressure is an incommensurate solid with zero point vacancies whose condensation leads to the formation of the SS. The theory can be used to address several phenomena observed in the PSU experiments and also make sharp predictions to be tested by possible future experiments, especially X-ray scattering experiments in the SS state.

2. Ginsburg-Landau theory of $^4$He: Let’s start by reviewing all the known phases in $^4$He. The density of a normal solid (NS) is defined as $n(\vec{x}) = n_0 + \sum_\xi n_\xi e^{i\vec{G} \cdot \vec{x}}$ where $n^*_\xi = n_{-\xi}$ and $\vec{G}$ is any non-zero reciprocal lattice vector. In a normal liquid (NL), if the static liquid structure factor $S(k)$ has its first maximum peak at $k_n$, then near $k_n$, $S^{-1}(k) \sim r_n + c(k^2 - k_n^2)^2$. If the liquid-solid transition is weakly first order, it is known that the classical free energy to describe the NS transition is

$$f_n = \frac{1}{2} \sum_\vec{G} |n_\vec{G}|^2 - w \sum_\xi n_\xi n_{\xi n} n_\delta \delta_{\vec{G}_1 + \vec{G}_2 + \vec{G}_3, 0} + u \sum_\vec{G} n_{\vec{G}_1} n_{\vec{G}_2} n_{\vec{G}_3} n_{\vec{G}_4} \delta_{\vec{G}_1 + \vec{G}_2 + \vec{G}_3 + \vec{G}_4, 0} + \cdots (1)$$

where $r_\vec{G} = r_n + c(G^2 - k_n^2)^2$ is the tuning parameter controlled by the pressure.

It was known that the Superfluid (SF) to Normal Liquid (NL) transition at finite temperature is a 3d XY transition described by:

$$f_\psi = K |\nabla \psi|^2 + t|\psi|^2 + u|\psi|^4 + \cdots (2)$$

where $\psi$ is the complex order parameter and $t$ is the tuning parameter controlled by the temperature.

The coupling between $n(\vec{x})$ and $\psi(\vec{x})$ consistent with all the symmetry can be written down as:

$$f_{\text{int}} = \frac{1}{2} v_1 n(\vec{x}) |\psi(\vec{x})|^2 + \cdots (3)$$

where the interaction must be repulsive $v_1 > 0$. Due to the lack of particle-hole symmetry in the NS, additional terms like $n(\vec{x}) \psi^4 \partial_\vec{x} \psi$ should exist and is very important at zero temperature and will be investigated in [11]. However, in the classical phase transitions investigated in this paper, this term can still be neglected. As shown in section 5, the repulsive interaction is proportional to the temperature shift $T_{SF} - T_{SS}$ in Fig.1.

In an effective GL theory, $n(\vec{x})$ and $\psi(\vec{x})$ emerge as two independent order parameters. The total density of the system is $n_\xi(\vec{x}) = n_0 + |\psi(x)|^2$ where $n(x)$ is the normal density and $|\psi(x)|^2$ is the superfluid density. A NS is defined by $n_\vec{G} \neq 0$, $\phi > 0$, while a SS is defined by $n_\vec{G} \neq 0$, $\phi = 0$. From the NL side, one can approach both the SS and the SF. Inside the NL, $t > 0$, $\psi$ has a gap, so can be integrated out, we recover the NS-NL transition tuned by $r_\vec{G}$ in Eqn.1 (Fig.1). Inside
the NL \( < n(\bar{x}) >= n_0 \), so we can set \( n_{\beta} = 0 \) for \( \vec{G} \neq 0 \) in Eqn.3, then we recover the NL to SF transition tuned by \( t \) in Eqn.2 (Fig.1).

Although the NL-NS and NL-SF transitions are well understood, so far, the SF-NS transition has not been investigated seriously. This transition may be in a completely different universality class than the NL-NS transition, because both sides break two completely different symmetry: internal global \( U(1) \) symmetry and translational symmetry. It is possible that the solid reached from the SF side is a new kind of solid than the NS reached from the NL side. In the following, incorporating quantum fluctuations into \( f_\psi \) (see Eqn.4) and considering the repulsive coupling between \( \psi \) and \( n \) sector in Eqn.3, we will determine the global phase diagram of \( ^4\text{He} \).

3. Two-component Quantum GL theory in the \( \psi \) sector: In this section, we develop a two-component Quantum GL theory in the \( \psi \) sector to replace Eqn.2 to describe the superfluid side of \( ^4\text{He} \). The superfluid is described by a complex order parameter \( \psi \) whose condensation leads to the Landau’s quasi-particles. Although the bare \( ^4\text{He} \) atoms are strongly interacting, the Landau’s quasi-particles are weakly interacting. The dispersion relation of superfluid state is shown in Fig.1b which has both a phonon sector and a roton sector. In order to focus on the low energy modes, we divide the spectrum into two regimes: the low-momentum regime \( k < \Lambda \) where there are phonon excitations with linear dispersion and high momentum regime \( |k - k_r| > \Lambda \ll k_r \) where there is a roton minimum at the roton surface \( k = k_r \). We separate the complex order parameters \( \psi(\bar{x}, \tau) = \psi_1(\bar{x}, \tau) + \psi_2(\bar{x}, \tau) \) into \( \psi_1(\bar{x}, \tau) = \int_0^\Lambda \frac{d^d k}{(2\pi)^d} e^{i\bar{k} \cdot \bar{x}} \psi(\bar{k}, \tau) \) and \( \psi_2(\bar{x}, \tau) = \int_{|k - k_r| < \Lambda} e^{i\bar{k} \cdot \bar{x}} \psi(\bar{k}, \tau) \) which stand for low energy modes near the origin and \( k_r \) respectively. For the notation simplicity, in the following, \( \int_{|k - k_r| <\Lambda} \) means \( \int_{|k - k_r| < \Lambda} \). The QGL action in the \( \psi \) sector in the \( (\bar{k}, \omega) \) space becomes:

\[
\mathcal{S}_\psi = \frac{1}{2} \int \left( \frac{d^d k}{(2\pi)^d} \sum_{\omega_n} (\omega_n^2 + t + Kk^2) |\psi_1(\bar{k}, i\omega_n)|^2 + \frac{1}{2} \int \left( \frac{d^d k}{(2\pi)^d} \sum_{\omega_n} (\omega_n^2 + \Delta_r + v_r (k - k_{r})^2) |\psi_2(\bar{k}, i\omega_n)|^2 + u \int d^d x d\tau |\psi_1(\bar{x}, \tau) + \psi_2(\bar{x}, \tau)|^4 + \cdots \right) \tag{4}
\]

where \( t \sim T - T_{SF} \) where \( T_{SF} \sim 2.17K \) is the critical temperature of SF to NL transition at \( p = 0.05 \text{ bar} \) and \( \Delta_r \sim p_c - p \) where \( p_c \sim 25 \text{ bar} \) is the critical pressure of SF to the SS transition at \( T = 0 \).

4. SF to SS transition and global phase diagram: In the SF state, the Feynman relation between the Landau quasi-particle dispersion relation in the \( \psi \) sector and the static structure factor in the \( n \) sector holds:

\[
\omega(q) = \frac{q^2}{2mS(q)} \tag{5}
\]

In the \( q \to 0 \) limit, \( S(q) \sim q, \omega(q) \sim q \) recovers the \( \psi_1 \) sector near \( q = 0 \). The first maximum peak in \( S(q) \) corresponds to the roton minimum in \( \omega(q) \) in the \( \psi_2 \) sector, namely, \( k_n = k_r \). As one increases the pressure \( p \), the interaction \( u \) also gets bigger and bigger, the first maximum peak of \( S(q) \) increases, the roton minimum \( \Delta_r \) gets smaller and smaller. Across the critical pressure \( p = p_c \), there are two possibilities (1) The resulting solid is an incommensurate solid, then \( < \psi >= 0 \) (2) The resulting solid is an incommensurate solid with vacancies even at \( T = 0 \) whose condensation leads to \( < \psi > \neq 0 \) \([1,2,7]\). Which case will happen depends on the strength of the repulsive coupling \( v_1 \) and will be analyzed in the next section.

Case (1) is trivial, the SS phase in Fig.1 does not exist. In the following, we only focus on case (2). From Eqn.1 and Eqn.4, we can see that \( n \) and \( \psi_2 \) have very similar propagators, so the lattice formation in \( n \) sector with \( n(x) = \sum_\mathcal{G} n_\ell e^{i\mathcal{G} \cdot \bar{x}} \) and the density wave formation in \( \psi_2 \) sector with \( < \psi_2(\bar{x}) >= e^{i\mathcal{Q}_m \cdot \bar{x}} \sum_{m=1}^P \Delta_m e^{i\mathcal{Q}_m \cdot \bar{x}} \) where \( \mathcal{Q}_m = k_r \) happen simultaneously. The \( \psi_2 \) sector alone is described by \( n = 2 \) component \( (d + 1, d) \) with \( d = 3 \) Lifshitz action \([10]\). The repulsive coupling in Eqn.3 \( v_1 > 0 \) simply shifts the DW by suitable constants along the three unit vectors in the direct lattice. These constants will be determined in the next section for different \( n \) lattices. Namely, the SS state consists of two inter-penetrating lattices formed by the \( n \) lattice and the \( \psi \) superfluid density wave (SDW).

Combining the roton condensation picture in this section with the results in section 2, we can sketch the following global phase diagram of \( ^4\text{He} \) in case (2).

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**Fig.1:** (a) The phase diagram in case (2). \( T \) controls thermal fluctuations, while \( P \) tunes quantum fluctuations. The SS exists only when the repulsive coupling \( v_1 \) in Eqn.3 is sufficiently small. Thick (thin) lines are 1st (2nd) order transitions. The critical temperatures of NL to SF and NS to SS transitions drop slightly as the pressure \( p \) increases because of the quantum fluctuations \([11]\).

(b) The separation of low (phonon) and high (roton) momenta regime in the SF.
5. The NS to SS transition: In this section, we approach the SS phase from the NS side and determine its lattice structure. In the NL, the BEC happens in the \( \psi_1 \) sector at \( k = 0 \), \( \psi_2 \) has a large gap and can be simply integrated out. In the NS, \( \psi \) stands for the vacancies due to large zero point quantum fluctuations [1,2,7]. Due to the \( n \) lattice formation, the mass of \( \psi_1 \) was increased to \( t + v_1^{NS} n_0 = T - T_{SS} \). Subtracting it from \( \psi_1^{NL} = t + v_1^{NL} n_0 = T - T_{SF} \) leads to \( T_{SF} - T_{SS} = n_0 \Delta v_1 \).

For simplicity, one can set \( v_1^{NL} = 0 \). If \( v_1 \) is larger than a critical value \( v > v_{c1} \), then \( T_{SS} \) is suppressed to zero. This is the case (1) discussed in the last section. The SS disappears from the phase diagram Fig.1. The resulting solid is a commensurate solid. In the following, we only focus on \( v_1 < v_{c1} \), so the SS state exists in the Fig.1 with \( T_{SS} > 0 \) which is the case (2) in the last section. The resulting solid is an in-commensurate solid with vacancies even at \( T = 0 \) whose condensation leads to the formation of the SS. Then the temperature shift \( \Delta T = T_{SF} - T_{SS} \) is an effective experimental measure of the repulsive coupling \( v_1 \) in Eqn.3. In the presence of the periodic potential of \( n(x) \) lattice, \( \psi \) will form a Bloch wave, the \( u \) self-interaction in the \( \psi \) sector in Eqn.4 will favor extended Bloch wave over strongly localized Wannier state. In principle, a full energy band calculation incorporating the interaction \( u \) is necessary to get the energy bands of \( \psi \). Fortunately, qualitatively physical picture can be achieved without such a detailed energy band calculation. In the following, substituting the ansatz

\[ \psi_1(\vec{x}) = a e^{i \theta_1}, \quad \psi_2(\vec{x}) = b e^{i \theta_2} \sum_{m=1}^{P} \Delta_m e^{i Q_m \cdot \vec{x}} \]

where \( Q_m = Q \) into Eqn.3, we study the effects of \( n \) lattice on \( \psi = \psi_1 + \psi_2 \). In order to get the lowest energy ground state, we must consider the following 4 conditions: (1) because any complex \( \psi \) (up to a global phase) will lead to local supercurrents which is costly, we can also take \( \psi \) to be real, so \( Q_m \) have to be paired as anti-nodal points. \( P \) has to be even (2) as shown from the Feynmann relation Eqn.5, \( Q_m, m = 1, \ldots, P \) are simply \( P \) shortest reciprocal lattice vectors. Then translational symmetry of the lattice dictates that \( \epsilon(\vec{K} = 0) = \epsilon(\vec{K} = \vec{Q}_m) \), \( \psi_1 \) and \( \psi_2 \) have to condense at the same time. (3) The point group symmetry of the lattice dictates \( \Delta_m = \Delta \) and is real (4) The repulsive interaction \( v_1 > 0 \) favors \( \psi(x = 0) = 0 \), so the Superfluid Density Wave (SDW) \( \rho = |\psi|^2 \) can avoid the \( n \) lattice as much as possible. It turns out that the 4 conditions can fix the relative phase and magnitude of \( \psi_1 \) and \( \psi_2 \) to be \( \theta_2 = \theta_1 + \pi, \Delta = a / P \), namely, \( \psi = a e^{i \theta} (1 - \frac{\pi}{2} \sum_{m=1}^{P/2} \cos(Q_m \cdot \vec{x}). \]

In this state, the crystal momentum \( \vec{k} \) is \( \vec{K} \) and the Fourier components are \( \psi(\vec{K} = 0) = a, \psi(\vec{K} = \vec{Q}_m) = -a / P \) which oscillate in sign and decay in magnitude. In principle, higher Fourier components may also exist, but they decay very rapidly, so can be neglected without affecting the physics qualitatively. In the following, we will discuss 4 common lattices \( sc, fcc, bcc, hcp \) respectively.

(a) \( P = 6 \): \( \vec{Q}_i, i = 1, 2, 3, 4, 5, 6 \) are the 6 shortest reciprocal lattice vectors generating a cubic lattice. The maxima of the DW \( \rho_{max} = 2a \) appear exactly in the middle of lattice points at the 8 points \( \vec{a} = \frac{1}{2}(\pm a_1 \pm a_2 \pm a_3) \). They form the dual lattice of the cubic lattice which is also a cubic lattice.

(b) \( P = 8 \): \( \vec{Q}_i, i = 1, \ldots, 8 \) form the 8 shortest reciprocal lattice vectors generating a \( fcc \) reciprocal lattice which corresponds to a \( fcc \) direct lattice. The field is \( \psi(\vec{x}) = a(1 - \frac{1}{2}(\cos Q_1 \cdot \vec{x} + \cos Q_2 \cdot \vec{x} + \cos Q_3 \cdot \vec{x} + \cos Q_4 \cdot \vec{x} + \cos Q_5 \cdot \vec{x} + \cos Q_6 \cdot \vec{x})) \), The local superfluid density \( \rho_{DW} \) appearing along any square surrounding the center of the cube such as \( (1/2, 0, 0) \) etc. and the centers of any cube such as \( (1/2, 1/2, 1/2) \).

(c) \( P = 12 \): \( \vec{Q}_i, i = 1, \ldots, 12 \) form the 12 shortest reciprocal lattice vectors generating a \( fcc \) reciprocal lattice which corresponds to a \( bcc \) direct lattice. The field is \( \psi(\vec{x}) = a(1 - \frac{1}{2}(\cos Q_1 \cdot \vec{x} + \cos Q_2 \cdot \vec{x} + \cos Q_3 \cdot \vec{x} + \cos Q_4 \cdot \vec{x} + \cos Q_5 \cdot \vec{x} + \cos Q_6 \cdot \vec{x})) \), The maxima of the DW \( \rho_{max} = 4a \) appear around any lattice point of the cube such as \( (1/2, 0, 0) \) (or \( 1/2, 0, 0 \), \( 1/2, 1/2, 1/2) \).

(d) Unfortunately, a spherical \( k_r = Q \) surface can not lead to lattices with different lengths of primitive reciprocal lattice vectors such as a \( hcp \) lattice. This is similar to the classical liquid-solid transition described by Eqn.1 where a single maximum peak in the static structure factor cannot lead to a \( hcp \) lattice [10]. Here we can simply take the experimental fact that \( n \) forms a \( hcp \) lattice without knowing how to produce such a lattice from a GL theory Eqn.1. Because for an idea \( hcp \) lattice \( c/a = \sqrt{3} \), an \( hcp \) lattice has 12 nearest neighbours, so its local environment may resemble that of a \( fcc \) lattice. We expect the physics (except the weak anisotropy of NCRI in the \( hcp \) lattice discussed in [11]) is qualitatively the same as that in \( fcc \) direct lattice.

Let’s look at the prediction of our theory on X-ray scattering from the SS. For simplicity, we take the \( sc \) lattice to explain the points. The other lattices will be discussed in [11]. For a lattice with \( j = 1, \ldots, n \) basis located at \( \vec{d}_j \), the geometrical structure factor at the reciprocal lattice vector \( \vec{K} \) is \( S(\vec{K}) = \sum_{j=1}^{n} f_j(\vec{K}) e^{i \vec{K} \cdot \vec{d}_j} \) where \( f_j \) is the atomic structure factor of the basis at \( \vec{d}_j \). The X-ray scattering amplitude \( I(\vec{K}) \) \( \sim |S(\vec{K})|^2 \). For the SS in the \( sc \) lattice, \( \vec{K} = \frac{2\pi}{a}(n_1 \hat{i} + n_2 \hat{j} + n_3 \hat{k}), \vec{d}_1 = 0, \vec{d}_2 = \frac{2 \pi}{a}(\hat{i} + \hat{j} + \hat{k}) \), then \( S(\vec{K}) = 1 + f(-1)^{n_1 + n_2 + n_3} \) for \( f \sim f_{max} \sim a^2 \). It is \( 1 + f \) for even \( \vec{K} \) and \( 1 - f \) for odd \( \vec{K} \). In fact, due to large zero-point motion, any X-ray scattering amplitude \( I(\vec{K}) \) will be diminished by a Debye-Waller factor. As shown in [11], the lattice phonon modes \( \vec{u} \) in \( \psi \) are locked to those of \( n \), so there is a common Debye-Waller factor \( \sim e^{-\frac{4}{3} k^2 u^2} \) for both even and odd \( \vec{K} \). We conclude that the elastic X-ray scattering intensity from the SS has an additional modulation over that of the NS.
modulation amplitude is proportional to the maxima of the superfluid density $\rho_s^{\alpha z} \sim a^2$ which is the same as the NCRI observed in the PSU’s torsional oscillator experiments. Unfortunately, so far, the X-ray scattering data is limited to high temperature $T > 0.8K > T_{SS}$ [12]. X-ray scattering experiments on lower temperature $T < T_{SS}$ are needed to test this prediction.

The results achieved in this section indeed confirm Fig.1 achieved from the roton condensation picture in the last section. A low energy effective action involving the superfluid phonon $\theta$, the lattice phonons $\vec{u}$ and their couplings will appear in a future publication.

6. The vortices in the SS: In the sectors of $\psi$, there are topological defects in the phase winding of $\theta$ which are vortices. At $T \ll T_{SS}$, the vortices can only appear in tightly bound pairs. However, as $T \rightarrow T_{SS}$, the vortices start to become liberated, this process renders the total NCRI to vanish above $T > T_{SS}$ in the NS state. In the SF phase, a single vortex energy costs a lot of energy $E_v^{SF} = \frac{\hbar^2 \xi_{SF}}{4 \pi m_a} \ln \frac{\xi_{SF}}{\xi_{SF}}$ where $m$ is the mass of $^3He$ atom, $R$ is the system size and $\xi_{SF} \sim a$ is the core size of the vortex. This energy determines the critical velocity in SF $\psi_c^{SF} > 30 cm/s$. In the SS state, because in the center of the SS vortex core, $\psi = 0$, so the vortices prefer to sit on a lattice site of the $n$ lattice. Because the long distance behavior of SS is more or less the same as SF, we can estimate its energy $E_v^{SS} = \frac{\hbar^2 \xi_{SS}}{4 \pi m_a} \ln \frac{\xi_{SS}}{\xi_{SS}}$. We expect the core size of a supersolid vortex $\xi_{SS} \sim 1/\Lambda > 1/k_F \sim a \sim \xi_{SF}$. So inside the SS vortex core, we should also see the lattice structure of $n$. In fact, we expect that $\xi_{SS}$ is of the order of the average spacing between the vacancies in the SS. It is interesting to see if neutron or light scattering experiments can test this prediction. Compared to $E_v^{SF}$, there are two reductions, one is the superfluid density, another is the increase of the vortex core size $\xi_{SS} \gg \xi_{SF}$. These two factors contribute to the very low critical velocity $\psi_c^{SS} \sim 30 \mu m/s$.

7. Discussions on PSU’s experiments: Although the NS to SS transition is in the same universality class as the NL to SF one [8], it may have quite different off-critical behaviours due to the SDW structure in the SS state. We can estimate the critical regime of the NS to SS transition from the Ginsburg Criterion $\gamma^2 \sim \xi_{SS}^2 \Delta C$ where $\Delta C$ is the specific jump in the mean field theory. Because of the cubic dependence on $\xi_{SS}$, large $\xi_{SS}$ leads to extremely narrow critical regime, the 3D XY critical behavior is essentially irrelevant, instead mean field Gaussian theory should apply. This fact can be used to address two experimentally observable effects of the He3 impurities. The first one was already observed by the PSU experiments and the second one is underway [13].
(1) The unbinding transition temperature $T_{SS}$ is determined by the pinning due to impurities instead of by the logarithmic interactions between the vortices. So the $^3He$ impurities effectively pin the vortices and raise the unbinding critical temperature $T_{SS}$. On the other hand, $^3He$ impurities will certainly decrease the superfluid density in both the $\psi_1$ and $\psi_2$ sector just like $^3He$ impurities decreases superfluid density in the $^4He$ superfluid.
(2) The mean field specific heat jump at $T = T_{SS}$ may be smeared by the presence of $^3He$ impurities.

8. Conclusions: The GL theory developed in this paper leads to a global and unified picture of $^4He$ physics at any temperature and pressure. If the repulsive coupling in Eqn.3 is sufficiently small, the SS state becomes a ground state at zero temperature. The solid at high pressure is an incommensurate solid with zero point fluctuations generated vacancies whose condensation leads to the formation of the SS. Assuming this is indeed the case, we investigate the SS state from both the SF and the NS side and find completely consistent description of the properties of the SS state. We found that the SF to the SS transition is a first order transition driven by the collapsing of roton minimum in the SF side, while the NS to SS transition is described by a 3d $XY$ model with much narrower critical regime than the NL to SF transition. The SS state is a uniform two-component phase consisting of a normal lattice plus a commensurate superfluid density wave (SDW). The SDW in the SS state leads to a modulation on the X-ray scattering intensity over that of the NS. The modulation amplitude is proportional to the NCRI observed in the PSU’s torsional oscillator experiments.

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