Density Response of the Warm Dense Electron Gas beyond Linear Response Theory: Excitation of Harmonics

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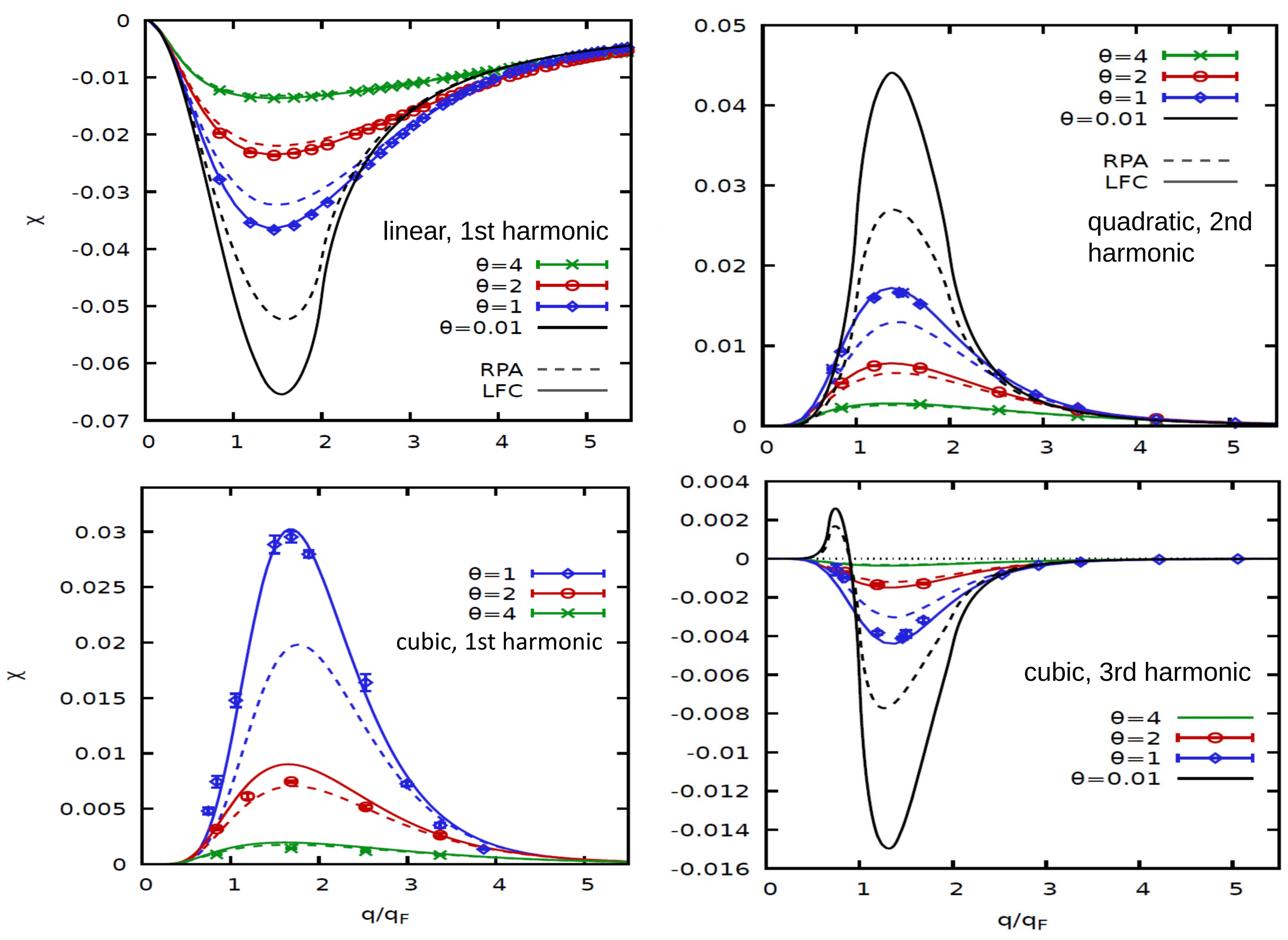
Abstract

Experimental diagnostics as well as theoretical modeling of warm dense matter (WDM) heavily rely on linear response theory. However, *Dornheim et. al.* [Phys. Rev. Lett.125, 085001 (2020)] showed that assuming the linear regime may not always be justified in experiments studying WDM. In addition, the intentional driving of non-linear effects should make new insight into many-particle effects possible. We use *ab initio* Path-Integral Monte-Carlo (PIMC) to obtain exact results for a harmonically perturbed homogeneous electron gas. A thorough analysis for different perturbation amplitudes is carried out. The corresponding density response reveals resonances at the higher harmonics of the perturbation wave vector. Analyzing the induced density response as a function of the perturbation amplitude shows the importance of the cubic response at the first harmonic and of the quadratic response at the second harmonic.



Nonlinear response, higher harmonics, comparison to theory

- temperature dependence well modelled by theory including LFC
- naturally, nonlinear influence vanishes quickly for limit of small q
- characteristic signatures in response functions for high degeneracy
- nonlinear responses allow for a more accurate interpretation of experiments where the weak-perturbation condition of LRT is violated

PIMC simulations of the perturbed electron gas

We use Path-Integral Monte-Carlo simulations of the uniform electron gas with a harmonic perturbation term

$$\hat{H} = \hat{H}_{\text{UEG}} + 2A\sum_{l=1}^{N}\cos(\hat{\mathbf{r}}_l\cdot\mathbf{q})$$

with N being the number of particles inside the simulation box and $\boldsymbol{q} = \frac{2\pi}{L}(n_x, n_y, n_z)^T$. We are interested in the density response. In order to achieve this, we compute the induced density

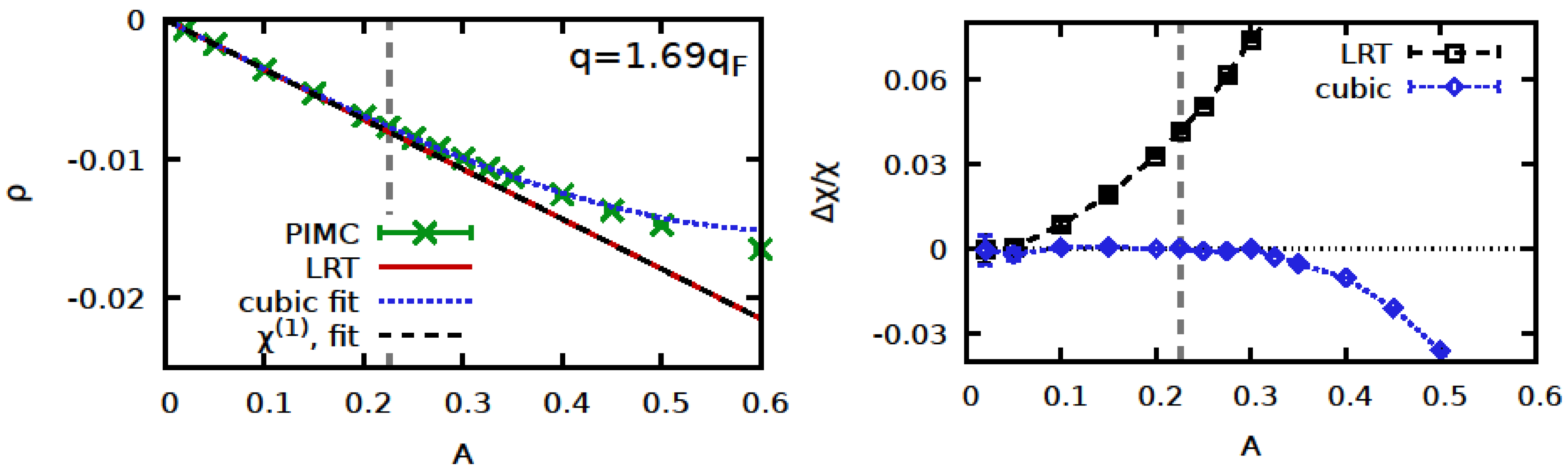
$$\langle\hat{\rho}_{\mathbf{k}}\rangle_{q,A} = \frac{1}{V}\left\langle\sum_{l=1}^{N} e^{-i\mathbf{k}\cdot\hat{\mathbf{r}}_l}\right\rangle_{q,A}.$$

The linear density response in PIMC can also be accessed using the imaginary-time version of the fluctuation-dissipation theorem.
In order to extract the nonlinear density response functions of various harmonics, we fit the induced density response to the expression

$$\langle\hat{\rho}_{\mathbf{q}}\rangle_{q,A} = \chi^{(1)}(q)A + \chi^{(1,cubic)}(q)A^3$$

with $\chi^{(1)}, \chi^{(1,cubic)}$ as free parameters. The cubic form fits the PIMC data very well and thus allows for the extraction of $\chi^{(1,cubic)}$. The simulations were performed with the following parameters

$$N = 14,\ r_s = \frac{\sqrt[3]{\frac{3}{4\pi n_e}}}{a_B} = 2,\ \theta = \frac{k_B T}{\varepsilon_F} = 1,\ q = \frac{2\pi}{L}(2{,}0{,}0)^T.$$



Nonlinear responses in the density profile

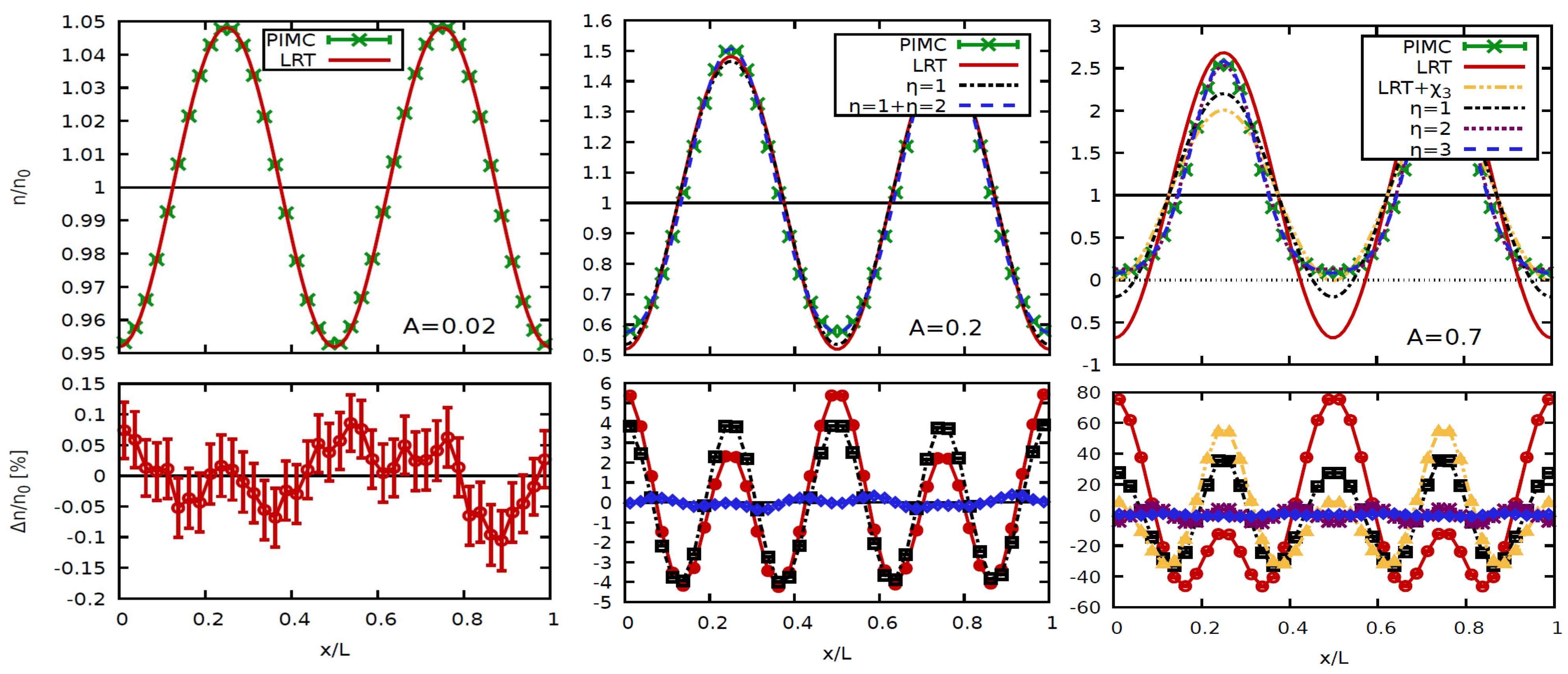
The right panels show the density profile in direction of the perturbation of the PIMC simulations with the same parameters as above. The red graphs depict the density profile in LRT

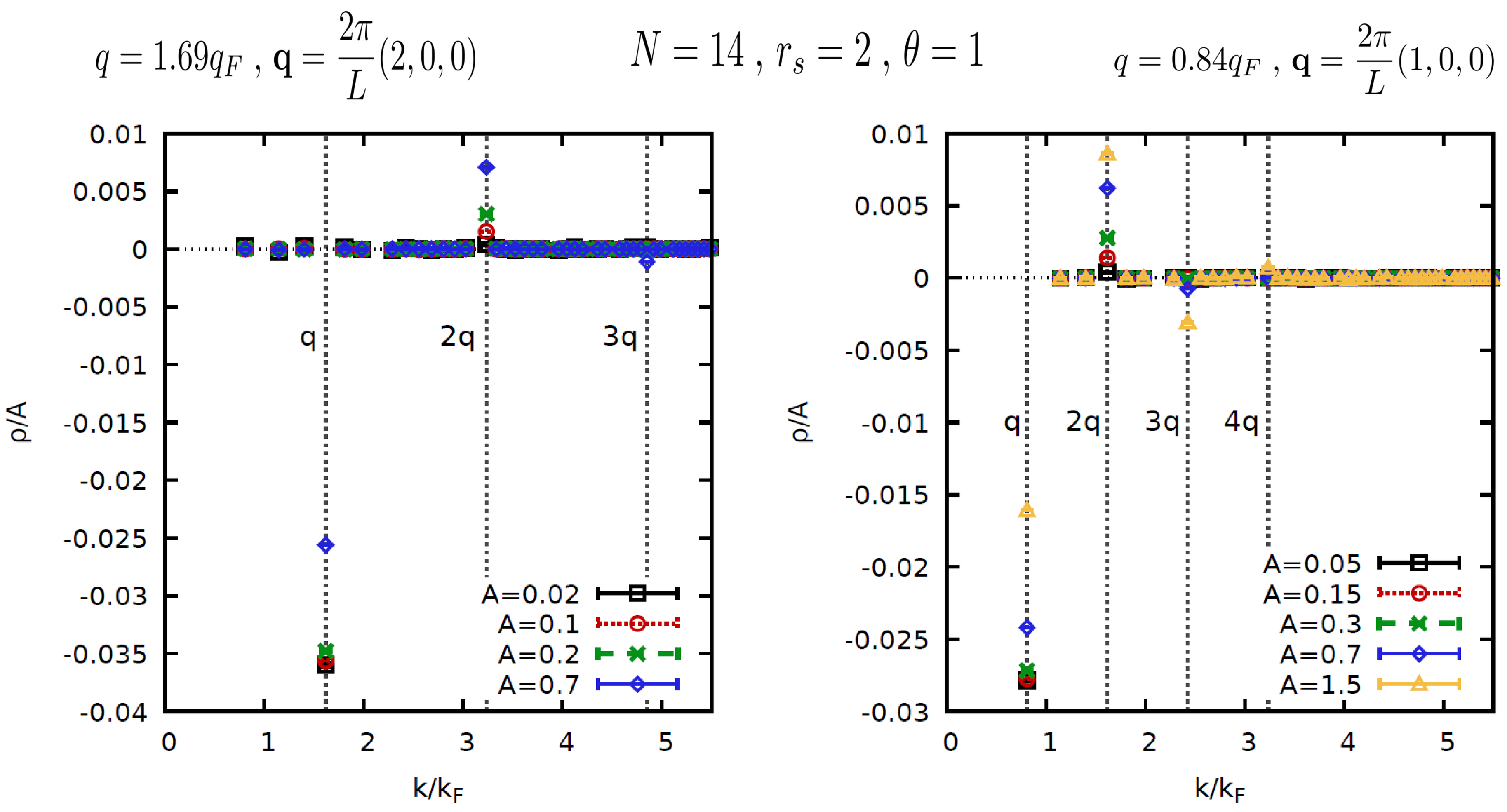
$$n(\mathbf{r}) = n_0 + 2A\cos(\mathbf{qr})\chi(\mathbf{q},0).$$

The non-linear contributions in the density profile is given by the expansion

$$n(\mathbf{r}) = n_0 + 2\sum_{\eta=1}^{\infty}\langle\hat{\rho}_{\eta\mathbf{q}}\rangle_{q,A}\cos(\eta\mathbf{q}\cdot\mathbf{r}).$$

The deviations between LRT and PIMC are especially visible at the maxima of the perturbation. With increasing A the higher harmonics start playing a more important role.





Nonlinear response calculation in PIMC, higher harmonics

- wave number resolved response for a static harmonic excitation for two different values of q as function of perturbation amplitude A
- quadratic response is the first order correction to linear response
- quadratic opposite sign (dampening of the excitation)
- third harmonic excitation well visible, even effects at fourth harmonic for large A
- drop of signal at first harmonic with increasing A due to influence of cubic response function

Outlook

Nonlinear effects play an important role when the basic assumption of a weak perturbation is not fulfilled anymore. This is especially the case for effects like nonlinear screening (effective potentials), stopping power, or coupled collective modes. Since nonlinear response is very sensitive to XC-effects, it can serve as a new diagnostic tool. The required conditions can be experimentally realized at XFEL facilities like the European X-FEL or LCLS.

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