# Femtosecond Response – Birth of a Quasiparticle


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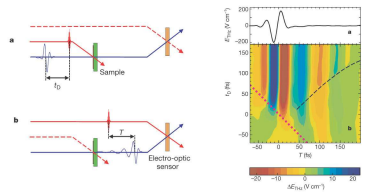
## Femtosecond laser irradiation




pump pulse creating charge carriers at time $t_0$ probe pulse time at time $t_D$ later

A. Leitenstorfer, Nature 426(2003)23, R. Huber et al. Nature 414(2001)286

Time and frequency–dependent dynamical response

$$\frac{1}{\epsilon(\omega,t)} = 1 + \frac{e^2}{\epsilon_0 q^2}\int_0^{t-t_0} dT e^{i\omega T}\chi(t,t-T)$$

time delay after probe pulse $T = t - t_D - t_0$

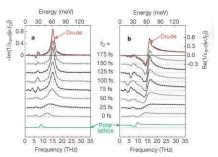
Fourier transformed into frequency

Quantum kinetic equations:

Gartner et al. PRB 66(2002) 075205

Vu and Haug PRB62 (2000) 7179

Kira and Koch PRL 93 (2004) 076402




## Solution of meanfield equation

• Linearize kinetic equation to obtain density response $\delta n(q,t) = \int_{t_0}^{t} dt' \chi(t,t') V_q^{\rm ext}(t')$

• one-particle reduced density matrix $\hat\rho$ obeys

$$\dot{\hat\rho} + i[\hat{\mathcal{E}} + \hat V^{\rm ind} + \hat V^{\rm ext}, \hat\rho] = \frac{\hat\rho^{\rm l.e.} - \hat\rho}{\tau}$$

• local equilibrium density matrix $\hat\rho^{\rm l.e.}$ deviates by chemical potential linked to $\delta n(q,t)$ by density conservation

• linearization leads to

$$\chi(t,t') = \Pi(t,t') + \int_{t'}^{t} d\bar t \left[\Pi(t,\bar t)\frac{e^2}{\epsilon_0 q^2} + I(t,\bar t)\right]\chi(\bar t,t')$$

• large-time limit $t+t' \to \infty$ approaches familiar form $\chi = \frac{\Pi^{\rm M}}{1-\frac{e^2}{\epsilon_0 q^2}\Pi^{\rm M}}$ with Mermin-Das polarization (Mermin PRB 1 (1970) 2362)

Solution of transient time response by one-sided Fourier transform

$$\frac{1}{\epsilon(\omega,t)} = 1 + \frac{e^2}{\epsilon_0 q^2}\int_0^{t-t_0} dT e^{i\omega T}\chi(t,t-T)$$

• limit of long wave lengths $q \to 0$ appreciable simplification $\Pi(t,t') \approx \frac{q^2 n(t')}{m}(t'-t)e^{\frac{t'-t}{\tau}}$ and $I(t,t') \approx \frac{1}{2}e^{\frac{t'-t}{\tau}}$

• integral equation into differential one with $n(t) \approx n_0\Theta(t-t_0)$

• Analytic solution Phys. Rev. B 72 (2005) 233203

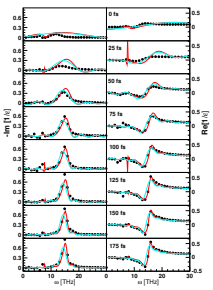
$$\frac{1}{\epsilon(\omega,t)} = 1 - \frac{\omega_p^2}{\sqrt{\omega_p^2 - \frac{1}{4\tau^2}}}\int_0^{t-t_0} dt' e^{(i\omega-\frac{1}{2\tau})t'} \sin\left(\sqrt{\omega_p^2 - \frac{1}{4\tau^2}}\,t'\right)$$

Achievement:

long-time limit yields the Drude formula $\lim_{t-t_0\to\infty}\frac{1}{\epsilon} = 1 - \frac{\omega_p^2}{\omega_p^2 - \omega(\omega+\frac{i}{\tau})}$

This limit is not so easy to achieve within short-time expansions, e.g. approximate result [ElSayed et al, PRB 49 (1994) 7337] gives $1 - \frac{\omega_p^2}{\omega_p^2 - (\omega+i/\tau)^2}$.

## Comparison with experiment




### GaAs

Resonance about 8 THz due to optical phonons, adding intrinsic contribution of crystal lattice

$\epsilon_{\rm GaAs} = \epsilon_\infty\left(\epsilon + \frac{\omega_{LO}^2 - \omega_{TO}^2}{\omega_{TO}^2 - \omega^2 - i\gamma\omega}\right)$

$\frac{\omega_{LO}}{2\pi} = 8.8$ THz, $\frac{\omega_{TO}}{2\pi} = 8.1$ THz lattice damping $\gamma$ =0.2 ps$^{-1}$ and background polarizability of ion lattice $\epsilon_\infty = 11.0$

Huber et al, phys.stat.sol.(b) 234 (2002) 207

pumb pulse at $t_0 = -40$fs, probe pulse full-width at half-maximum $27fs$, plasma frequency $\omega_p$ =14.4 THz, relaxation time $\tau = 85$ fs

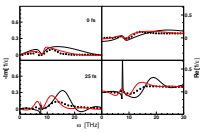
Improvement by realistic shape

• too fast build-up of the collective mode at the time $t = 25$ fs

• just the time duration of the experimental pulse

• we have approximated this by the instant jump

• more realistic smooth populating can be modeled by an arctan-function and numerical solution improves the description




First two time evolution steps comparing an instant population of conduction band with a smooth transition.

## If no meanfields ? → Levinson equation

Time diagonal part of Kadanoff/Baym equation $\Delta_E = \frac{k^2+p^2-(k-q)^2-(p+q)^2}{2m}$

$$\frac{\partial \rho_k}{\partial t} = \frac{2}{\hbar^2}\sum_{pq} V_q^2 \int_0^t d\bar t \cos\left\{\frac{t-\bar t}{\hbar}\Delta_E\right\}\left(\rho'_{k-q}\rho'_{p+q}(1-\rho_p)(1-\rho_k) - \rho_p\rho_k(1-\rho'_{k-q})(1-\rho'_{p+q})\right)$$

time-dependent Fermi's Golden Rule and memory effects

Properties:

• density conservation $\frac{\partial}{\partial t}n = 0$ • momentum conservation $\frac{\partial}{\partial t}\langle p_1\rangle = 0$

• Time variation of kinetic energy $\frac{\partial}{\partial t}(\langle\frac{p_1^2}{2m}\rangle + \langle V_{12}\rangle) \equiv 0$

Phys. Lett. A 199 (1995) 241 , Europ. J. Phys. A (1999) 291-305, Phys. Rev. C 64 (2001) 024613

The inclusion of memory effects produces the full energy conservation.

But, several effects hidden:

Off-shell tails in $\rho$, renormalization of scattering rates and wave function, quasiparticle energies collision delays Ann. Phys. (NY) 294 (01) 135

Better: neglect memory effects, only time-dependent Fermi's Golden Rule: e.g. Maxwellian plasma [Phys. Lett. A 246 (1996) 311; Phys. Rev. E 63 (2001) 20102]

static Debye $z = \omega_p\sqrt{t^2 - it\frac{\hbar}{T}}$:

$$\dot E_{\rm corr}(t) = -\frac{e^2 n\kappa T}{2\hbar}{\rm Im}\left[(1+2z^2)e^{z^2}(1-{\rm erf}(z)) - \frac{2z}{\sqrt{\pi}}\right]$$

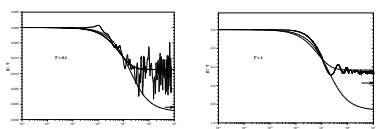
dynamical screening $z_1 = \omega_p\sqrt{2t^2 - it\frac{\hbar}{T}}$:

$$\dot E_{\rm corr}^{\rm dynam}(t) = -\frac{e^2\kappa T}{\hbar}{\rm Im}\left[e^{z_1^2}(1-{\rm erf}(z_1))\right]$$

classical limit

$$E_{\rm corr}^{\rm static}(t) = -\frac{1}{4}\kappa e^2 n\left[1 + \frac{2\omega_p t}{\sqrt{\pi}} - (1+2(\omega_p t)^2)e^{(\omega_p t)^2}(1-{\rm erf}(\omega_p t))\right]$$

$$E_{\rm corr}^{\rm dynam}(t) = -\frac{1}{2}e^2 n\kappa\left\{1 - \exp\left(\frac{\omega_p^2}{2}t^2\right)\left[1-{\rm erf}(\frac{\omega_p}{\sqrt{2}}t)\right]\right\}$$



$\Gamma = \frac{2}{7\pi}$

Comparison with MD simulations [G. Zwicknagel ]

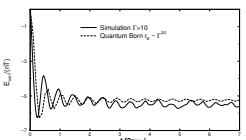
## Dense classical plasma by quantum Born

*On a correspondence between classical and quantum particle systems,* Phys. Rev. E 66 (2002) 022103

Quantum-Born [Europ. J. Phys. A (1997) 291]:

$$\frac{E_{\rm corr}^T(t) - E_{\rm corr}^0(t)}{nT} = \frac{1}{(36\pi^4)^{1/6}}\frac{r_s^3}{\Gamma}\left(\frac{\sin y\tau}{y\tau} - 1\right)\left(\frac{1}{b_t}\arctan(\frac{1}{b_t}) + \frac{1}{b_t^2 + b_t^4}\right)$$

Bruckner parameter $r_s = \frac{d}{a_B}$, $b_t = \frac{\hbar\kappa}{2\mu} = \sqrt{\Gamma}(48\pi^2)^{-1/6}$, time in plasma periods $\tau = \frac{2\pi}{\omega_p}t$ or $y\tau = 4\epsilon_f t/\hbar = (2)^{4/3}\pi^{5/3}3^{5/6}\tau/\sqrt{r_s}$



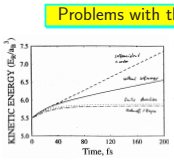

Simulation of dense classical plasma [G. Zwicknagel]

best fit ($\hbar \sim T$):

$$r_s^{\rm fit} = \frac{1}{2}\sqrt{\frac{3}{8}}\Gamma^{3/2}$$

$$\lambda^2 = \frac{d}{\kappa} = \frac{d^2}{(3\Gamma)^{1/2}} \approx d^2$$

## Problems with the Levinson equation



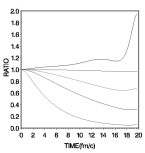

Continuous increase of kinetic energy D. Kremp, M. Bonitz, W.D. Kraeft, M. Schlanges: Ann. of Phys. 258 (1997) 320

Numerical instability L. Banyai, D. B. T. Thoai, C. Remling, H. Haug: Phys. Stat. Sol. (B) 173 (1992) 149

Pulsation modes S. L. Popyrin: Dokl. Phys. 43 (1998) 671

Two-time correlation function from Kadanoff/Baym equation $G^<(\mathbf{p},t,t')/G^<(\mathbf{p},t,t)$ vs $t-t'$ for four different strengths of interaction and $\mathbf{p} = 0$. Uppermost curve is from Levinson calculation.

Phys. Rev. C 64 (2001) 024613




Problem: Double count of correlations(retardation)

Solution: Extended quasiparticle picture Ann. of Phys. 294 (2001) 135

## Summary

• collisions have no time to happen yet (only meanfields are formed)

• separate gross feature of the formation of collective modes at transient times which are due to mean-field fluctuations

• simple analytic formula for time dependence of the dielectric function

• subtracting this gross feature from the data allows to extract effects due to higher-order correlations, have to be simulated by quantum kinetic theory beyond the mean field

But: relaxation time needed and what if no meanfields (quasi-neutrality) ?

• Short time behavior by time-dependent Fermi's golden rule ≡ Finite duration approximation of non-Markovian collision integrals
  -low temperature value is universal; ratio $\sim \hbar/\epsilon_F$ (formation of quasiparticles), high temperature limit $\sim 1/\omega_p$
  -analytical results for dynamical screening, static screening leads to 1/2 correlation energy
  -good agreement with solution of Kadanoff and Baym equation

• Initial correlations induce a current and energy contribution (over/under-correlated initial state leads to decrease/increase of kinetic energy)

• later times ≡ higher order correlations: Nonlocal kinetic equation
  -Unifies Landau - quasiparticle picture for quantum liquids and Enskog - like equations for dense gases
  -Thermodynamically consistent equation of state including quantum 2. virial coefficient
  -Memory effects result in:
  – Off-shell tails of Wigner function (exactly compensated)
  – Renormalization of scattering rates
  – Collision delay is all what is left from memory
  – Related non-local corrections to the scattering integral
  -successfully applied: Heavy Ion collisions, deep neutral impurities, Bernoulli potential at superconducting surfaces