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COHERENCE OF ELECTRON PHOTOEXCITATION BY EXTREMELY SHORT STRONG LIGHT PULSES

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For a semiconductor alloy with a predominant disorder scattering, we show that, under appropriate conditions, electrons photoexcited by a short strong light pulse form a coherent transient at first, while the incoherently backscattered electrons take over only gradually, with a time lag comparable with the pulse duration. The time evolution of the electron distribution is obtained by a direct evaluation of the non-equilibrium Green function.

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1. Introduction

Our present aim is to study the electron photoexcitation in semiconductors, involving time evolution of the band populations and of the transient polarization in the so-called coherent regime, when the decohering or randomizing processes set on with a certain time lag, and so they have a minor effect during the pulse [1], for a recent example see [2]. This advance has been made possible by a steady reduction of the pulse durations into the femtosecond range, that is to durations shorter than most of the coherence times, even in systems as fluid as the free electron-hole pairs in semiconductors. As a result, coherent transients and the onset of the decohering scattering processes can be separated by varying the pulse used.

While this physical picture is clear, a systematic theoretical description is not easy. Up to now, the time delay of these chaotization processes has been interpreted mostly in a qualitative fashion against an intuitive background. This intuitive approach can be given a more systematic basis using non-equilibrium Green functions (NGF), which permit a simultaneous rigorous treatment of the coherent and of the incoherent excitation. In fact, the NGF structure itself leads to an incoherent part of the excitation which appears to set on with a time lag, but persist. By contrast, the coherent part follows in a synchronous manner the exciting pulse, but it is purely transient.

A direct determination of the NGF is commonly considered as prohibitively difficult, and at present a systematic approximate approach is in general use, introducing quasiparticles into the propagators and using factorization called generalized Kadanoff-Baym ansatz (GKBA) [3] to reduce the dynamics of the particle correlation function $G^<(t, t')$ to the semiconductor optical Bloch equations for the density matrix $\rho(t)$ [4, 5].

We should like to argue that this approach has considerable weaknesses, if we choose to study a situation favorable for the coherent regime. Then, all characteristic times, the pulse duration and its Rabi period (reciprocal to its strength), and the electron coherence time(s) become comparable, ≈ 100 fs. In the absence of a characteristic time hierarchy, it appears as important to avoid approximate constructs, like quasiparticle states, which are based on such hierarchy [6].

Fortunately, among the scattering electron mechanisms, there is one, namely the *elastic scattering on a random alloy/impurity potential*, for which the task of a direct computation of the NGF is not hopeless, as proposed in Ref. [7]. The way from the proposal to its accomplishing was not easy. Here, we report for the first time the results of a full direct calculation of the non-equilibrium Green function for a light pulse driven and strongly non-equilibrium photoelectron transient in a semiconductor alloy.

2. Model and its treatment using the two-time non-equilibrium Green functions

The simple model we use is a two-band semiconductor alloy introduced in Refs [7, 8]. The system considered resembles a direct gap semiconductor of the (Ga,Al)As-type, with disorder acting only in the conduction band. The initial condition is then non-random, because all electrons start in the full valence band. The choice of a coherent pulse with a ground frequency high compared to the pulse duration permits to use the rotating wave approximation (RWA), which makes the model equivalent to a transient light induced hybridization of crossed bands [9]. The alloy case is convenient because of an easy variability by a mere variation of the composition of the alloy, which allows us to adjust the parameters of the problem in a wide range. In particular, the alloy scattering can be made dominant for very short times.

The one-electron Hamiltonian will be decomposed into the mean-field and fluctuating parts as

$$H(t) = H_{\text{MF}}(t) + \mathcal{D}, \quad H_{\text{MF}}(t) = \langle H(t) \rangle, \quad \langle \mathcal{D} \rangle = 0. \quad (1)$$

Here, $H_{\text{MF}}(t)$ consists of the virtual crystal Bloch Hamiltonian and of the coupling to the light pulse, which makes it time dependent. The configuration averages are denoted by $\langle \dots \rangle$.

The quantity of the direct physical interest is the electron distribution (one-electron density matrix). It is obtained as

$$\rho(t) = -i\hbar G^<(t, t) \quad (2)$$

from a double-time *particle correlation function* $G^<(t, t')$. This rather complex quantity has the advantage that it obeys closed equations of motion, the so-called

Dyson equations [10, 11]. It should be warned, however, that the only way of calculating the photoexcitation dynamics for a pulse of a general shape is to solve these equations in the time domain. Here, we may only sketch, how this can be achieved in practice.

For the particle correlation function $G^<$, we have, if ρ_0 denotes the non-random initial state one-electron density matrix (occupied valence band),

$$G^<(t, t') = i\hbar G^R(t, t_0)\rho_0 G^A(t_0, t') + \int_{t_0}^t \int_{t_0}^{t'} dt_1 dt_2 G^R(t_1, t_2) \Sigma^<(t_1, t_2) G^A(t_2, t'). \quad (3)$$

Here, G^R and G^A are the averaged electron propagators describing the electron coherent waves. Their Dyson equations were solved in Ref. [6]. The two terms in Eq. (3) describe respectively *the coherent part and the incoherent part of the particle correlation function*. A similar separation follows for the density matrix (2).

To obtain, from Eq. (3), a closed equation of motion, we have to make a physical approximation by expressing the self-energy $\Sigma^<$ as an approximate functional of $G^<$. Presently, we use the self-consistent Born approximation,

$$\Sigma^<(t, t') = \langle \mathcal{D}G^<(t, t') \mathcal{D} \rangle. \quad (4)$$

To generate $G^<(t, t')$, we found as the easiest way to solve an equivalent integro-differential equation. The details will be reported elsewhere.

3. Numerical results

Figure 1 represents the total photoexcitation per unit cell $\text{Tr}\rho_{cc}$ from the full valence band to an empty conduction band for three different sech^{-2} pulses, which are sketched by vertical hatches. Band and pulse parameters are given in the figure caption. In the upper left corner we see the effect of the "weak long" pulse. Both parts of the excitation are comparable. The incoherent part persists, while the coherent one is purely transient. This is not a universal feature, and it is due to heavy damping of the coherent propagator in the present case.

The "strong short" pulse (lower left), whose strength is increased so as to cause the same asymptotic excitation as the first pulse, demonstrates the feasibility of achieving of a nearly complete coherence of the excitation during the pulse. Finally, for the "strong long" pulse (upper right), the net excitation increases. An important physical feature are the Rabi oscillations (RO). They are a visible manifestation of the transient and rather elusive "Galitskii gap". Apparently, it opens up for three RO periods. Interestingly enough, the incoherent excitation increases in three RO like waves to a final value, which is roughly three times larger than in the previous case. In the SCBA we used, the RO for coherent and the incoherent part coincide in phase, as predicted in Refs. [12, 13], because the shift due to virial corrections (*collision delay*) is negligible.

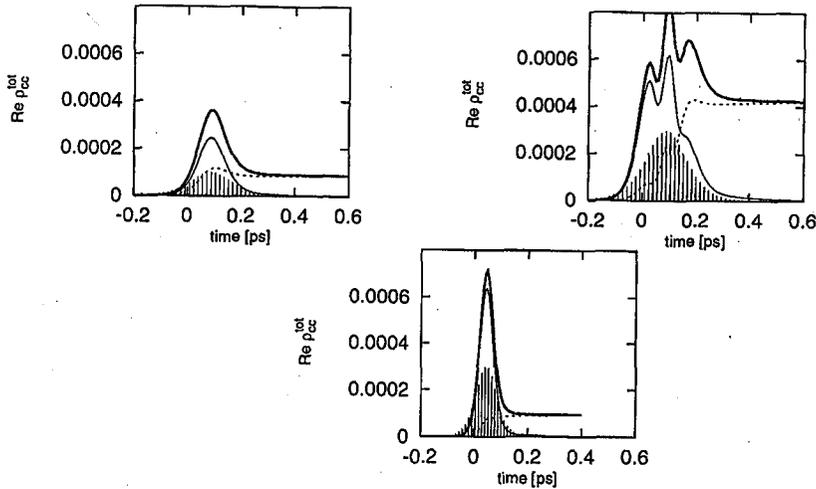


Fig. 1. The total photoexcitation from the full valence to an empty conduction band ($\text{Tr}\rho_{cc}^{\text{tot}}$) for three different pulses. The total photoexcitation (heavy line) is split into two components (see Eq. (3)), coherent part — thin line, incoherent part — dashed line. The pulse shape $\propto [\exp(t/a) + \exp(-t/a)]^{-2}$ indicated in an arbitrary vertical scale (by vertical hatches). The resonant photon energy in all three cases is $\hbar\omega = 2$ eV. Upper left corner: $a = 0.10$ ps (halfwidth = 0.175 ps), field hybridization strength $Q = 0.01$ eV; upper right corner: $a = 0.10$ ps (halfwidth = 0.175 ps), $Q = 0.03$ eV; lower left corner: $a = 0.05$ ps (halfwidth = 0.088 ps), $Q = 0.02$ eV; photon frequency in all three cases — 2.0 eV. Band and disorder parameters as in Ref. [6]: $E_G = 1.5$ eV, $m_c^* = 0.4$, $m_v^* = 0.6$, alloy scattering strength = 0.03 eV.

4. Conclusion

Our main aim was to demonstrate that for the coherent spectroscopy using femtosecond pulses, the NGF technique may be the right theoretical partner. The numerical example shows that pronounced coherent effects (Rabi oscillations) may be observable in the free electron-hole gas even if the electron coherence time is as short as $10^1 \div 10^2$ fs. By tailoring the pulse in a rather restricted range, substantial variations in the coherence of the electron response can be achieved. These predictions open new paths for designing the coherent fs spectroscopy experiments.

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