Analytical formulation for electron excitation in dielectrics under an intense laser field

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Electron excitation in dielectrics by an intense laser field is the main process in laser–matter interactions. Technical developments in femtosecond laser processing have made it possible to produce a nanoscale laser-induced periodic surface structure (LIPSS), and to realize nonthermal ablation for subwavelength resolution [1–4]. For femtosecond lasers, electron excitation by multiphoton ionization and tunnel ionization is crucial, because such nonlinear processes generate a controllable free-carrier density and confine the material change to the focal volume. Therefore, the prediction of the electron excitation rate using theoretical models and/or numerical simulation is important.

Keldysh proposed a theory for the rate of electron excitation under an intense linearly polarized laser field [5]. His approach is very general and can be used to describe the photoionization of different objects from single atoms to crystals. Because of its generality, the Keldysh model has attracted much attention and has become one of the standard tools in the theory of laser photoionization. For atoms and molecules, the Keldysh–Faisal–Reiss (KFR) theory, which is an implementation of the original Keldysh work, is one of the most important theories in understanding the electron–laser interaction.

Jones and Reiss have developed a formula for the rate of electron excitation under a circular polarized laser employing the *S*-matrix theory [6]. Whereas the Keldysh formula treats the time-dependent wavefunction of the valence and conduction bands as the Houston function and includes only the reduced electron–hole mass, the Jones and Reiss formula treats only the conduction band as the Houston function (Volkov state) and includes the effective mass of the valence and conduction bands independently. Therefore, a direct comparison between the Keldysh and Jones formulas is not relevant.

The purpose of this work is to construct an analytical formula for the transition probability in dielectrics including multiphoton and tunneling processes under a circularly polarized laser. We derive the transition probability in a crystalline solid under a circularly polarized laser, assuming a parabolic two-band system and using the Houston function [7] as the time-dependent wave function.

The total transition probability induced by the circularly polarized laser is found to be

$$W_c = \frac{e^2A_0^2P^2\mu^{3/2}}{\sqrt{2}\pi m^2c^2}\sum_{l=l_0}^{\infty}\int d\theta \sin\theta \left(J_{l-1}^2(\eta') + J_{l+1}^2(\eta')\right)\sqrt{\zeta_l}, \quad (1)$$

where

$$\eta' = \frac{eA_0\sqrt{2\zeta_l}\sin\theta}{\sqrt{\mu}\omega c}, \quad (2)$$

and

$$\zeta_l = l\omega - (\mathrm{B}_g + U_c). \quad (3)$$

Here, $J_l(\eta)$ is the Bessel function.

The total transition probability induced by the linearly polarized laser is found to be

$$W_L = \frac{e^2A_0^2P^2\mu^{3/2}}{2\sqrt{2}\pi m^2c^2}\sum_{l=l_0}^{\infty}\int d\theta' \sin\theta' \left(J_{l-1}(\alpha,\beta) + J_{l+1}(\alpha,\beta)\right)^2\sqrt{\kappa_l}, \quad (4)$$

where

$$\kappa_l = l\omega - \left(B_g + U_p\right). \quad (5)$$

Here, θ' is the angle between the polarization direction and $\vec{k}$, and l_0 is the maximum integer l at which $\kappa_l > 0$. $J_l(\alpha,\beta)$ is the generalized Bessel function and $U_p = e^2A_0^2/4\mu c$ is the ponderomotive energy. α and β are defined as

$$\alpha = \frac{eA_0\sqrt{\kappa_l}\cos\theta'}{\sqrt{\mu}\omega c}, \beta = \frac{e^2A_0^2}{8\mu\omega c^2}. \quad (6)$$

α-quartz is a typical dielectric used in nonlinear laser–matter interaction studies, and we selected it here as an example with which to illustrate the application of our developed formalism. The transition probability (W_L) of α-quartz by linearly polarized 800-nm light is shown in Fig. 1 by a red solid line.

We assumed a band gap of 9 eV and reduced mass of $0.30m$ [8]. W_L calculated by the full expression of the conventional Keldysh formula (dashed line) and tunneling limit (dot-dashed line) are also shown in Fig. 1 for comparison. Our formalism shows excellent agreement with the Keldysh theory. This result indicates that our formula includes the multiphoton and tunneling processes as the Keldysh formula does.

The transition rate induced by circularly polarized 800-nm light as a function of laser intensity is illustrated in Fig. 2 as a blue dashed line. The transition rate induced by linearly polarized 800-nm light is also shown as a solid red line. The power law of the transition rate induced by the circularly polarized light is slightly different from that for linearly polarized light, and higher than that for linearly polarized light of higher intensity. Temnov *et al*. [9] reported the ratio of the ionization rate around an intensity of 1×10^{13} W/cm^2. From the relationship between the

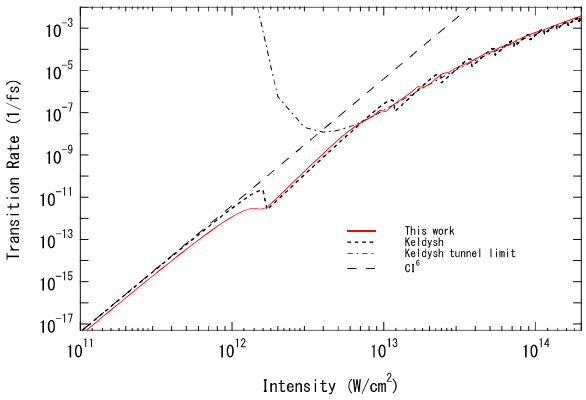

Fig. 1 Transition probability as a function of laser intensity for linearly polarized 800–nm light in α-SiO$_2$. The red solid line represents the excitation rate determined by our formalism, the blue dashed line represents the excitation rate based on the full expression of the Keldysh theory, the green dotted line represents the tunneling limit of the Keldysh theory, and the black dot-dashed line represents the simple six-photon process.

intensity in vacuum (I_v) and that in media (I_m), $I_m = \varepsilon^{1/2} I_v$,

where ε is the dielectric constant, 1×10^{13} W/cm^2 corresponds to 4×10^{12} W/cm^2 in Fig. 2. The experimental value for the excitation rate ratio W_c/W_L is 0.3. Our result gives ratios of 0.1–0.2 at (2×10^{12})–(1×10^{13}) W/cm^2, which is in reasonable agreement with the experimental value.

We extended the Keldysh-type formula for the solid state under an intense circularly polarized laser, assuming the Houston function for the valence and conduction bands. Because our formula depends only on the reduced mass, it can be directly compared with the Keldysh formula. Our simple formula describes the electron excitation rate, reproduces the Keldysh formula with excellent agreement for α-quartz, and makes it possible to separate the contribution of each l-photon process with linear or circular polarization. The ratio of the rate of transition under linear polarization to that under circular polarization determined using our formula shows reasonable agreement with the experimental results.

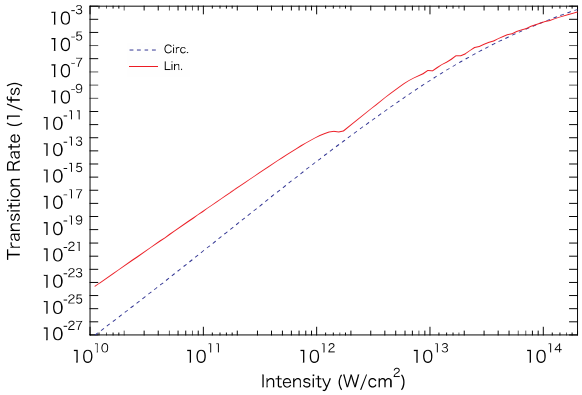

Fig. 2 Transition probability as a function of electric field intensity for linearly polarized (red solid line) and circularly polarized (blue dashed line) light.

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