Strong field processes inside gallium arsenide

S M Golin\textsuperscript{1,2}, S E Kirkwood\textsuperscript{1}, D D Klug\textsuperscript{3}, D M Villeneuve\textsuperscript{3}, D M Rayner\textsuperscript{3}, C A Trallero Herrero\textsuperscript{1,2} and P B Corkum\textsuperscript{1}

\textsuperscript{1}Joint Attosecond Science Laboratory, National Research Council and University of Ottawa, 100 Sussex Drive, Ottawa ON K1A 0R6, Canada
\textsuperscript{2}Physics Department, Kansas State University, Manhattan, KS 66506, USA
\textsuperscript{3}National Research Council, 100 Sussex Drive, Ottawa ON K1A 0R6, Canada

E-mail: kirkwood@ieee.org, trallero@phys.ksu.edu and Paul.Corkum@nrc-cnrc.gc.ca

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Abstract

We demonstrated experimentally that the multiphoton ionization rate in gallium arsenide depends on the alignment of the laser polarization with respect to the crystal axis. We observed modulation in the ionization rate of a linearly-polarized 1900 nm laser beam directly by measuring its transmission while rotating the crystal, without Fourier analysis. We propose that the modulation in the ionization rate arises from periodic variation in the reduced carrier mass, as predicted by Keldysh theory. We show direct comparison of the experimental transmission modulation depth with Keldysh’s non-resonant ionization theory for solids. This opens up a novel method for non-invasive crystallography of semiconductor materials.

Keywords: strong fields, ultrafast, ionization, GaAs, semiconductors

(Some figures may appear in colour only in the online journal)

1. Introduction

Fifty years ago, Keldysh published his theory on multiphoton ionization (MPI) that laid the foundation for strong field physics [1]. His atomic theory of ionization has been extensively studied [2] and refined [3, 4], leading to significant advances in our understanding of modern strong field phenomena in atomic and molecular systems such as attosecond pulse generation [5–8], high harmonic generation [9–13] and ‘scanning tunnelling microscopy’ of orbital structures [14].

Keldysh treated atoms and solids in a similar manner. Considering ‘ionization’ in a solid as the transition of an electron from the valence band to the conduction band, we can see the direct analogy between an atomic orbital and the band structure of a solid. By defining what we now call the ‘Keldysh parameter’, $\gamma$, MPI in these two systems were delineated between a tunnelling limit where $\gamma < 1$, and a perturbative limit when $\gamma > 1$. Much of modern strong field physics in atoms and molecules occurs on this dividing line where $\gamma \approx 1$.

Despite the advancements in our understanding of atomic and molecular multiphoton processes over these past decades, a similar revolution in our understanding of this process in solids has not occurred. The challenge in studying this process in solids is largely due to difficulties in isolating it from the complexity of interactions between a strong field and a high density of atoms in a periodic structure. Immediate experimental differences also arise when moving from atoms to solids since the products of ionization, such as electrons, ions, or high-harmonic generation, remain confined within the bulk. Several groups have studied ultrafast dynamics in solids in the weak field limit [15–18], but very little theoretical and experimental work has been done with strong fields. Early work in the strong field regime has been done in dielectrics [19, 20].

We can exploit the significantly higher density of electrons in solids ($10^{22}$ cm$^{-3}$ versus $10^{10}$ cm$^{-3}$ in gases) to develop experiments that measure changes in the laser beam itself, specifically, the fractional change in transmission of the laser propagating through the material. Depletion of the laser beam due to nonlinear absorption gives an indirect measure of the ionization probability. Similar experiments on MPI in solids measuring changes in transmission were performed with dielectrics by Gertsvo\textit{f et al} [19]. In these experiments,
large bandgap crystals were ionized with a linearly-polarized, 800 nm laser beam and a periodic variation in the transmission was observed as a function of crystal rotation. The observed periodicity matched the symmetry of the crystal, which was linked to differences in the MPI rate.

Keldysh’s theory on MPI in solids is dependent on the effective carrier mass, directly related to band curvature near the minimum bandgap. The theory assumed a parabolic band structure, requiring a constant and definitive effective mass. Despite the more complicated band structures in the dielectrics tested, those experiments showed promising qualitative results. Moreover, it has been recently shown that light-induced currents in dielectrics can be created with strong fields [20]. With the observation of similar atomic strong field processes in semiconductors, such as high harmonic generation [21, 22], interest has increased in Keldysh’s treatment of solids. The recent developments in longer wavelength femtosecond lasers into the mid-IR [23] now allow us to study semiconductors where new photonic devices can be developed. These lasers also allow us to isolate this ionization process from linear, resonantly nonlinear and thermal processes, and test for more accurate theoretical predictions, as was done with atoms and molecules these past decades.

In this manuscript, we show that strong-field ionization of solids near the tunneling limit can be used to trace the crystal symmetry and reveal interband absorption dynamics near the bandgap of solid periodic structures. By rotating a GaAs crystal about a linearly-polarized, infrared femtosecond laser beam, we measured periodic variations in the fractional transmission and compared it to predictions based on Keldysh’s MPI model for solids. Our comparisons are further aided by more accurate predictions of the effective mass through simulations using density functional theory (DFT).

2. Experimental setup

To measure the crystal structure of GaAs using Keldysh’s theory for solids, we selected the idler output from a superfluorescence-seeded fs-TOPAS optical parametric amplifier (OPA) made by Light Conversion, which was pumped by a regeneratively-amplified titanium-doped sapphire (Ti:S) laser. The Ti:S was a homemade system having a center wavelength of 800 nm, which produced 50 fs full-width at half-maximum (FWHM) pulses at a repetition rate of 500 Hz with a maximum pulse power of 800 μJ. The linearly-polarized OPA idler beam was adjusted to produce 1900 nm (0.6 eV), 75 fs (FWHM) pulses.

A schematic of our experimental setup is shown in figure 1. The OPA output was filtered by a set of dielectric mirrors to select the idler beam followed by a 600 μm thick GaAs crystal set at the Brewster angle (73°) to filter out any residual wavelengths created through additional nonlinear processes within the OPA.

The idler beam was passed through an electronically-controlled shutter to control the exposure time for each crystal angle. A half-wave plate and polarizer were used to vary the incident pulse energy, and the angle of the half-wave plate was also automated to control the input energy of the idler beam. A photodiode (PD1) was calibrated against the incident pulse energy, measured by a power meter placed at a reference plane between the lens and the sample to account for Fresnel losses from the lens surfaces, as illustrated by the dotted line in figure 1.

In gas ionization theory, the ionization threshold approximately scales with the ionization potential as $I_{th} \propto \frac{I}{P^4}$, independent of the wavelength. Treating the bandgap in a solid as analogous to the ionization potential, we estimated a minimum intensity above which to measure ionization. In dielectrics with a bandgap of 9 eV, the ionization threshold was approximately $10^{13}$ W cm$^{-2}$. Scaling down to the 1.43 eV
The bandgap of GaAs, an ionization threshold of approximately $10^{10} \text{W cm}^{-2}$ was predicted. Our pulse energy on the target could vary from 0.9 $\mu$J to 6 $\mu$J, with a 1.1 mm incident beam radius ($1/e$) as measured using a knife-edge, our intensity range varied from $3 \times 10^{10}$ to $2 \times 10^{11} \text{W cm}^{-2}$ when focussed with a 200 mm focal length plano-convex lens.

The sample was mounted on a rotation stage which itself was mounted on a two-dimensional translation stage to allow us to select any spot on the surface. In addition, a kinematic mount was attached to the rotation stage to align the sample for normal incidence. The laser transmission through the sample was collected with an integrating sphere to ensure the entire beam was collected. Traces from both the transmission and reference photodiodes were processed with a boxcar integrator and sent to a data acquisition card. This setup allowed for single shot measurements synchronized to the laser shutter.

3. Crystal properties

At a wavelength of 1900 nm, the semi-insulating GaAs(100) with a bandgap of 1.43 eV was transparent, giving negligible linear absorption. GaAs is not birefringent when light propagates along a principal axis and should not induce any additional symmetry dependence. Since these experiments involved at least three-photon absorption, we can disregard all two-photon absorption effects. Finally, because of the high electron carrier mobility, the electrons quickly replenish between laser pulses.

GaAs has a face-centred cubic (FCC) zinc-blende structure as shown in figure 2. The structure space group is F$\bar{4}3$ m. Since it is a cubic structure, it has a four-fold symmetry and the angles of the unit cell are all at 90°. Figure 2 is viewed along the (001) plane, which can be seen as parallel to the $i-j$ axes and orthogonal to the $k$ axis. The (100) plane is identical to the (010) and (001) planes.

The first Brillouin zone is shown in figure 2. The $\Gamma$ point is at the origin with zero momentum and the boundary sits at a momentum of $k_i = 2\pi/a$. The high symmetry points ($\Gamma$, $X$, $W$, $L$) can be visualized more clearly in reciprocal space and all lie on the boundary. As expected, the GaAs reciprocal lattice has a four-fold symmetry, analogous to the GaAs real-space lattice. Due to the direct relationship between the spaces, the laser can be visualized as propagating along direction, $k_i$, where $i$ can be any of $x$, $y$, or $z$.

For our experiments, we need to know the valence and conduction band curvatures along the laser polarization direction as the crystal is rotated about the propagation axis. We modeled the GaAs band structure around the axis of rotation using ABINIT [24], a DFT with pseudopotential functions. Figure 3 shows the band structure for the first section of the Brillouin zone (from $X$ to $W$). The band structure can be envisioned along paths directing in and out from the central point, $\Gamma$; first to point $X$ and then to incrementing points, $Z_i$ in increasing angles about the propagating axis, $k_i$. Points $Z_i$ are initially defined in reciprocal space and

![Figure 2](image1.png)

![Figure 3](image2.png)
then converted into real space to find the corresponding real-space angle. Similar curves were made for the rotation from \( W \) to \( W' \), and \( W' \) to \( X' \). Due to the four-fold symmetry, the pattern repeats in each quadrant.

The effective mass of a carrier in a periodic potential is dependent on the curvature of the band as shown in the following equation [25]:

\[
m^* = \frac{\hbar^2}{\epsilon} \left( \frac{d^2 \epsilon}{dk^2} \right)^{-1},
\]

where \( \epsilon \) is the band energy and \( k \) is the lattice vector. The second derivative of the band structure at the \( \Gamma \) point for each angle was calculated numerically from the DFT data using a least-squares fitting algorithm, the Savitzky–Golay method [26].

Since ionization involves the transition of a carrier from one energy surface to the next, the overall mass of the carrier being ionized is the reduced mass, defined as,

\[
\frac{1}{m^*} = \frac{1}{m^*_v} + \frac{1}{m^*_c},
\]

where \( m^*_v \) and \( m^*_c \) are the effective masses of the valence band and conduction band, respectively. The absolute values were used since the sign of the mass infers directionality of the carrier acceleration.

The reduced mass for ionization from either the heavy hole or light hole valence band is shown in figure 4. The light hole reduced mass varies with a smaller amplitude than the heavy hole mass and the phase of the two curves differ by 90°. As the amount of laser transmission is expected to be inversely related to the reduced mass [19], one would expect a transition from a maximum to a minimum if ionization dominantly occurred from the heavy hole valence band, with the opposite occurring if the light hole valence band dominated.

4. Experimental results

When the sample was rotated in a plane normal to the laser propagation direction, the transmission was measured for each angle, \( \theta \) as shown in figure 5. The angle, \( \theta \), is defined as the sample’s angle with respect to the polarization axis of our linearly-polarized laser beam, where 0° corresponds to alignment of the [001] axis with the laser polarization.

The transmission, \( T \), was defined as

\[
T = \frac{E_{out}}{E_{in}} = \frac{E_{in} - E_{abs}}{E_{in}},
\]

where \( E_{abs} \), \( E_{in} \), and \( E_{out} \) are the energy absorbed, input pulse energy, and output pulse energy, respectively.

In addition to transmission, we defined a modulation depth by first fitting a \( A + B \sin^2(2\theta) \) curve to the experimental data using MATLAB’s least square fit, where \( A \) and \( B \) are fitting parameters. The modulation depth was defined as the difference between the maximum and minimum transmission. These curves were reproduced for the laser energy range discussed in the experimental section.

The least square error between the modulation data and the \( \sin^2(2\theta) \) fit is approximately 2%, which is similar to the pulse-to-pulse fluctuations of the laser. From this fitting
defined as [1]

$$\gamma = \omega \sqrt{\frac{E_g m^*}{eF}}.$$  \hspace{1cm} (4)

where $\omega$, $E_g$, $m^*$, and $F$ are the angular frequency, bandgap energy, effective mass, and field strength, respectively. This Keldysh parameter for solids differs from the atomic case by a factor of $1/\sqrt{2}$.

In the tunnelling regime, the transition rate can be written in terms of $\gamma$ as,

$$w_{\text{Tunnel}}(\gamma) = \frac{2}{9\pi} \frac{E_g}{\hbar} \left( \frac{m^* E_g}{\hbar^2} \right)^{3/2} \left( \frac{\hbar \omega}{E_g} \right)^{5/2} \Phi \left( \frac{1}{\gamma^2} \right) \left( 1 - \frac{1}{1 - \gamma^2} \right)^{(x+1)}.$$ \hspace{1cm} (5)

From this equation, we see that the tunnelling regime begins to break down when $\gamma > 2.8$ (as the intensity decreases). As $\gamma \gg 1$, the rate enters the perturbative multi-photon regime (MPI) and the expression becomes,

$$w_{\text{MPI}}(\gamma) = \frac{2}{9\pi} \omega \left( \frac{m^* E_g}{\hbar^2} \right)^{3/2} \Phi \left( \frac{2(x + 1) - 2x^{1/2}}{1 - 4\gamma^2} \right) \left( 1 - \frac{1}{16\gamma^2} \right)^{(x+1)}.$$ \hspace{1cm} (6)

where $x = \Delta/\hbar \omega$, $\Phi$ is the Dawson integral, the brackets $\langle \rangle$ represents the integer portion, and $\Delta$ is the effective bandgap energy. In the MPI regime, the effective bandgap can be written as,

$$\Delta = E_g + \frac{\omega^2 F^2}{4 m^* e^2},$$ \hspace{1cm} (7)

where the second term in this expression is equivalent to the ponderomotive energy.

In these sets of experiments, the intensity range spans both the MPI and tunnelling regimes necessitating the use of the full theory. Using the following simplifying expressions:

$$\gamma_l = \frac{1}{\gamma^2 + 1},$$
$$E_l = E(\gamma_l) \quad E_\gamma = E(\gamma\gamma_l), \quad K_l = K(\gamma_l), \quad K_\gamma = K(\gamma\gamma_l),$$ \hspace{1cm} (8)

where $E(\gamma_l)$ and $K(\gamma_l)$ are elliptic integrals of the first and second kind, and generalizing the effective bandgap for all values of $\gamma$ as

$$\Delta = \frac{2E_l E_\gamma}{\pi\gamma_l},$$ \hspace{1cm} (9)

which recovers the MPI limit in equation (7), we can rewrite
the Keldysh model for solids, equation (37) in [1], as

\[
\gamma (\omega) = \frac{2\omega}{9\pi} \left( \frac{m^* \omega}{\hbar f} \right)^2 Q(\gamma, \frac{\Delta}{m^* \omega}) \times \exp \left\{ -\pi \left( \frac{\Delta}{\hbar \omega} + 1 \right) \right\} K_x - E_v, \tag{10}
\]

where the function \( Q \) is

\[
Q(\gamma, x) = \sqrt{\frac{\pi}{2K_1}} \sum_{n=0}^{\infty} \exp \left\{ -\pi n \frac{K_x - E_v}{E_1} \right\} \times \phi \left( \frac{\pi^2 (2(x + 1) - 2x + n)}{2K_1 E_1} \right) \tag{11}
\]

and the index, \( n \), indicates the multiphoton order. Note that the effective bandgap \( \Delta \) increases with increasing laser intensity, which we refer to as channel closing as the intensity of the laser increases.

Figure 6 shows the ionization rate for GaAs at 1900 nm extending a bit beyond our intensity range using a reduced mass of 0.06877 \( m_e \) (heavy hole transition at 0°). The figure shows that ionization is effectively modeled using the perturbative MPI approximation at low intensities (high \( \gamma \)), equation (6). As \( \gamma \) drops below 2.8, with increasing intensity, we approach the tunnelling regime and the tunnelling approximation in equation (5) is valid. The full Keldysh MPI model in solids, equation (10), shows a sharp dip related to channel closing as the effective bandgap increases and four photons are required to span the gap rather than three. The tunnelling approximation does not include this effect, and although the perturbative approximation includes channel closing, it models it at a lower intensity with an overall lower ionization rate, which justifies using the full model in this transition region between approximations where \( \gamma \approx 1 \).

### 6. Discussion

In figure 7, the transmission of the 1900 nm laser beam for a fixed incident polarization angle of 0° was plotted normalized to the transmission of a linearly-absorbed laser beam (accounting for Fresnel losses). The curves in figure 7 were calculated by propagating an initially Gaussian temporal and spatial intensity distribution through a 600 \( \mu \)m thick sample and depleting the simulated pulse using the space–time averaged Keldysh ionization rate with the reduced mass between the lowest conduction band and the heavy hole valence band, for the blue curve, and the light hole valence band for the dashed red curve.

The energy absorbed by the material in each time step, \( dE_{abs} \), is given below,

\[
dE_{abs} = \eta(w(I)N)dt,
\]

with \( w(I) \) being the space–time averaged Keldysh ionization rate for a given reduced mass, and \( N \) being the electron number density.

Although the independent laser parameters: pulse energy, pulse width, and incident beam waist, were measured and should lead to an accurate calculation of the incident intensity, the experimental and the theoretical transmission data were still offset in intensity. We measured the pulse width of the 800 nm laser to be 50 fs (FWHM), but a corresponding measurement of the idler pulse width was not possible. We estimated the pulse would stretch to about 75 fs from 50 fs based on similar OPA pulse measurements. Furthermore, the
knife-edge measurement of the incident beam indicated
additional structure to the beam, possibly increasing the \( M^2 \)
above one, leading to a larger focal spot. For these initial
experiments, we did not attempt to clean the spatial mode.

In gas ionization and high-harmonic generation, similar
variabilities in the prediction of the laser intensity from the
independent measurements and the focal spot conditions have
been observed [27, 28]. As in those experiments, we used the
transmission curve to assess the experimental intensity of the
laser at focus inside the sample, similar to how the ionization
threshold calibration was used in gas-phase strong field
experiments. Energy loss due to Fresnel reflection off the
front and back sample surfaces was corrected using the
reflection coefficient of GaAs at 1900 nm. Since the phase of
the modulation curve seen in figure 5 confirms that the heavy
hole valence electrons are predominantly ionized, fitting the
experimental data to the heavy hole curve in figure 7 is further
justified.

The transmission decreases as the laser pulse intensity
increases because the beam is depleted by absorption of the
laser energy required for carrier band-to-band transitions. The
steep decline in transmission as a function of intensity shows
evidence of nonlinear absorption, since linear absorption
would only depend on the sample thickness and not the pulse
energy. The hump seen in the experimental data at ~5 \( \times \) \( 10^{10} \)
W cm\(^{-2} \) is not repeatable and thus part of the experimental
error. These experiments were repeated and this current data
set was chosen for completeness.

Figure 8 shows the modulation depth as a function of
intensity inside the sample using the intensity as calibrated by
the transmission curve of figure 7. The points represent the
experimental data and the solid blue and dashed red lines are
the theoretical transmission curves for the heavy and light
hole band ionization, respectively. The large fluctuation seen
in the experiment around 5 \( \times \) \( 10^{10} \) W cm\(^{-2} \) is due mainly to
power fluctuations from the laser propagating through the
OPA.

The theoretical curves show an initial increase in mod-
ulation depth as the ionization rate is angle-dependent.
Figure 5 shows that there is little ionization at \((2n + 1)\pi/4\) degrees compared to ionization at \(n\pi/2\) degrees, where
\(n \in \mathbb{Z} \). As the intensity continues to increase, the modulation
depth plateaus when ionization at \((2n + 1)\pi/4\) degrees is no
longer negligible. At much larger intensities, the modulation
depth decreases as the pulse is depleted of photons, showing
less variation between transmission along different sample
angles.

Given the high intensity and short pulse width of the
laser pulse, other nonlinear effects could result in transmission
variations as explored here, in particular self-focusing and
phase advance. In the case of self-focusing, the beam
would experience an increase in the refractive index from the
intensity-dependent refractive index. For GaAs, the nonlinear
index is \( n_2 = 3.3 \times 10^{-13} \) cm\(^2\) W\(^{-1} \). Self-focusing would
overcome the beam’s divergence if its power was above a
critical power, [29], which is 4.8 kW for 1900 nm propagating
through GaAs(100) crystal.

Rather than stay below the critical power, we used a
relatively large beam size so that the change in beam waist
due to self-focusing would be negligible. We chose a focusing
lens of 200 mm, which gives a predicted beam waist of 110
\( \mu \)m and a peak power greater than 1 MW. The Rayleigh range
was 2 cm, which is over 30 times the thickness of the sample.

Using the maximum energy output of the 1900 nm laser
system (~40 \( \mu \)J), the distance over which the beam would
collapse due to self-focusing was calculated to be 1.4 mm
[29]. Using simple geometry, the output intensity would
increase by a factor of 1.6, purely due to self-focusing.

As shown in [30], however, ionization occurs at the peak
of the laser as it approaches the focal spot, depleting and
clamping the peak intensity. Using equation (10) and the
ionization simulations described previously, we found that the
intensity falls to approximately 1/6 times its maximum value
upon exiting the sample, limiting the effect of self-focusing.

Phase advance may occur when the refractive index is
decreased due to plasma generation in the material, causing
the beam to diverge (or self-defocus). As the material is
ionized by an incoming electric field, the carrier density
(plasma) will increase, reducing the refractive index. We used
the classical electron oscillator model (Drude model for
dielectrics and semiconductors) to find the change in index to
be,

\[
\Delta n(\lambda) = -\frac{e^2 \lambda^2 \Delta N}{8\pi^2 n_0 \varepsilon_0 c^2 m},
\]

where \(N\) is the free electron (plasma) density in cm\(^{-3}\), and \(m\) is
the effective mass, and \(n_0\) is the refractive index.

We estimated that the minimum carrier density needed to
achieve a phase advance of \(\pi\) to be 9.66 \( \times \) \( 10^{15} \) cm\(^{-3}\) for
1900 nm. For an input Gaussian beam, the input laser fluence
is given by \(\sim I_{FWHM} \). The fluence removed from the beam
through ionization to reach a phase advance of \(\pi\) is \(\sim N_e E_e L\),
assuming a constant interaction region over the full length of
the sample. We estimated the minimum transmission for
which the phase advance becomes significant by taking the
ratio of remaining fluence in the beam over the input laser
fluence. We found that for 1900 nm pulses, the minimum
transmission is 0.56. Using our Keldysh model, we find that
this transmission value is not reached until the peak intensity
exceeds \(14 \times 10^{10} \) W cm\(^{-2} \), which was not possible in our
experiments. The lowest transmission achieved in the
1900 nm experiments was \(\sim 0.75\) meaning that phase advance
was negligible.

The experimental data again shows that Keldysh ion-
ization from the GaAs heavy hole valence band predicts the
ionization rate. Although the ionization rate of the light hole
valence band is greater than that for the heavy holes, the
heavy hole valence band shows a much greater variation in
reduced mass as a function of angle. The ionization rate from
the heavy hole band varies by 33.7% while the light hole band
varies by only 8.7% (Both depths are calculated theoretically
for a peak intensity of \(10 \times 10^{10} \) W cm\(^{-2} \) and averaged over the
laser spot).
The transmission experiments used to measure crystal symmetry in dielectrics by Gertsvolf et al. [19] also used the Keldysh ionization rate. Using DFT to produce the band structure for various dielectrics, they showed that the reduced mass was angle-dependent when using a linearly polarized, 800 nm laser beam on a rotated sample. With parabolic fitting at the band minimum for direct bandgap dielectrics, the ionization was shown to be most probable between the top of the valence band to the bottom of the conduction band, as shown here, due to the exponential-dependence on the bandgap. In that work, they discovered that the difference in the reduced mass between two symmetry points, a factor of 2 in quartz, was not a predictor in the difference in the ionization rates. They measured a difference of about 1% and required Fourier analysis to visualize the crystal symmetry. As seen in figure 5, the modulation depth for GaAs varied from 2% to 5%, which allowed us to reveal the crystal symmetry directly without the need of Fourier analysis. This difference in modulation depth for GaAs results from a much smaller variation in our reduced masses between our symmetry points than seen in dielectrics.

Further evidence of ionization from the heavy hole valence band results from calculating the density of states for each band from our DFT results. In the $\Gamma$–X to $\Gamma$–W directions, the conduction band DOS decreases from $2 \times 10^{17} \text{cm}^{-3}$ to approximately $1.5 \times 10^{17} \text{cm}^{-3}$ at W. As for the heavy hole valence band, there is a degeneracy of 2 and the band responsible for modulation used in this work has a DOS that increases from $4.7 \times 10^{17} \text{cm}^{-3}$ to $8.8 \times 10^{17} \text{cm}^{-3}$. The light hole valence band decreases from $1.1 \times 10^{17} \text{cm}^{-3}$ to $0.7 \times 10^{17} \text{cm}^{-3}$. Although the ionization rate from the light hole valence band should be higher given the smaller reduced mass, the conduction band would not be filled by the ionized light valence electrons and the higher density of heavy hole valence electrons would dominate ionization.

Also observed in Gertsvolf et al. [19], transmission variations with crystal symmetry in quartz vanished when the photon energy was doubled using 400 nm laser pulses. The absence of this variation was attributed to the ionization transitioning into the perturbative regime, where $\gamma > 1$. The variation in transmission was still observed in LiF, though, and this variation was attributed to the ionization still being in the tunnelling regime with four-photon absorption. In our work, we are not in a purely tunnelling regime as shown plotted to the right in figure 6 and ionization only required three photons for absorption as the effective bandgap would not exceed this energy until the intensity increased beyond $40 \times 10^{10} \text{W cm}^{-2}$. Although we were in a three-photon absorption regime where ionization could not be said to purely tunnelling, a significant variation in transmission was observed counter to that in observed in quartz at 400 nm by Gertsvolf et al. The complexity of the band structure in quartz, as compared to LiF and GaAs here, may be a factor, providing more incentive to explore Keldysh’s ionization theory in solids with different crystal structures, especially as longer wavelength lasers become available.

7. Conclusion

We demonstrated that our experimental technique confirms theoretical predictions of MPI inside bulk GaAs using Keldysh’s non-resonant MPI theory for solids. This evidence was revealed as a decrease in the transmission of 1900 nm pulses as a function of laser intensity. In addition, we experimentally show that the MPI (band-to-band excitation) of GaAs(100) reveals the crystal’s symmetry. This symmetry was observed by rotating the laser polarization with respect to the crystal axis and measuring the laser transmission. We observed a distinct modulation in the transmission that repeated every 90°, congruous to the four-fold symmetry of a GaAs cubic fcc crystal. Prediction of semiconductor crystal symmetry followed similar results seen by Gertsvolf et al. [19] with dielectrics. The modulation depth was more significant than seen in any previous work, allowing visualization of crystal symmetry without further Fourier analysis.

The experimentally measured transmission and the modulation depth were predicted by Keldysh’s non-resonant MPI theory for solids. In this theory, ionization depended on the reduced mass, which results from the curvature of the band structure along different directions of laser polarization. We showed that the modulated transmission results primarily from ionization between the heavy hole valence band and the conduction band at the direct bandgap.

Fifty years after the publication of Keldysh’s theory of MPI and the significant advances made in his atomic theory for atoms and molecules in predicting modern strong field phenomena, these experiments and the development of longer mid-IR wavelength lasers will open the way to similar advances in strong field ionization in solids.

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