

Article

Constant Matrix Element Approximation to Time-Resolved Angle-Resolved Photoemission Spectroscopy

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Abstract: We discuss several issues associated with employing a constant matrix element approximation for the coupling of light to multiband electrons in the context of time-resolved angle-resolved photoemission spectroscopy (TR-ARPES). In particular, we demonstrate that the “constant matrix element approximation” —even when reasonable—only holds for specific choices of the one-electron basis, and changing to other bases, requires including nonconstant corrections to the matrix element. We also discuss some simplifying approximations, where a constant matrix element is employed in multiple bases, and the consequences of this further approximation (especially with respect to the calculated TR-ARPES signal becoming negative). We also discuss issues related to gauge invariance of the final spectra.

Keywords: pump/probe photoemission; time-resolved angle-resolved photoemission spectroscopy; constant matrix element approximation; nonequilibrium many-body physics; gauge-invariance

1. Introduction

Time-resolved and angle-resolved photoemission spectroscopy (TR-ARPES) is emerging as one of the powerful tools in ultrafast optics used to probe the non-equilibrium behavior of quantum materials and how they recover back to a quasiequilibrium state. Many different materials have been investigated so far, including charge-density-wave systems [1–9], high-temperature superconductors [10–14], iron pnictides [15], Mott insulators [16], and topological insulators [17]. Theory has also been well developed to model these systems. The theory developed by Freericks, Krishnamurthy, and Pruschke [18] described the most general formulas for this pump/probe spectroscopy (including corrections to ensure gauge invariance and including the dominant second-order perturbation theory contributions [19]). Because the formula for TR-ARPES (in a gauge) comes from second-order perturbation theory, it involves the square of a many-body matrix element, which, in general, can depend on the band index, momentum parallel to the surface, the energy of the photoelectron (see Equation (7) below) and time, and can be shown to be explicitly nonnegative. The true physical response, however, is the gauge-invariant response, and it is not hard to show that the general formula with the exact matrix elements is indeed gauge invariant. However, as we discuss below, this can be a problem when approximations are made for the matrix elements, and while a gauge invariant prescription can be constructed following a straightforward procedure [20], it has proved to be difficult to establish, in general, that this result is non-negative [19].

It is also common to invoke a “constant matrix element approximation”, wherein its dependence on its various arguments is entirely neglected, within theoretical calculations of photoemission spectra. This is not necessarily because the matrix elements are known to be constant, but because explicitly determining the matrix elements is extremely challenging for quantum materials, so employing a constant matrix element is a good first step in modeling such systems. The constant matrix element approximation is trivial when there is only a single effective band in the system, but it becomes more complex when there are multiple bands. In this paper, we discuss different options one has for invoking the constant matrix element approximation within TR-ARPES, paying particular attention to the issues of gauge invariance and non-negativity of the spectra. We also discuss implications for time-resolved photoemission spectroscopy (TR-PES), where the momentum dependence has been integrated over.

2. Results

The description of a pump/probe experiment is complicated by the fact that the presence of the pump removes time-translation-invariance symmetry from the system, and we generally find that the Green’s function depends on two times. Using the Wigner coordinates [21], these two times can be summarized as the average time $t_{\text{ave}} = (t_1 + t_2)/2$ and the relative time $t_{\text{rel}} = t_1 - t_2$. The pump must be explicitly included into the Hamiltonian, because it is a large “perturbation” of the system, so it cannot be treated with low-order perturbation theory. Fortunately, because the Peierls substitution [22] allows us to treat the electric-field effects exactly for single-band models, we have an exact means to include the effects of the pump pulse. Note that in multiband models, there can be additional electric field effects due to symmetry-allowed dipole transitions that connect two bands that have opposite spatial parity (and, of course, higher-order multipoles can also be taken into account, but it is rarely needed to do so). The dipole approximation would involve a direct coupling (hybridization) between the two bands, with the strength of the coupling proportional to the field amplitude. This can also be easily included into the Hamiltonian, which then fully describes the effects of the electric field (however, magnetic field effects—including fields generated by time-varying electric fields—are usually neglected). The other constraint is that the field is uniform in space. Non-uniform fields create many additional complications to the formalism. The probe pulse, on the other hand, is usually taken to be weak, and hence it can be treated with lowest order (second-order) perturbation theory. While there are a number of different second-order terms that can contribute, only one term dominates, and that is the only one we will focus on here [18,19].

2.1. Formalism for the TR-ARPES Response in a Gauge

It was demonstrated in reference [18] that the TR-ARPES signal can be found from a probe-pulse-weighted two-time Fourier transform of the non-equilibrium lesser Green’s function. We need to review that derivation to first correct it for a proper gauge-invariant treatment, and to establish our notation so that we can discuss the constant matrix-element approximation.

We assume the Hamiltonian $\mathcal{H}(t)$ includes all of the interactions of the electrons (with themselves, and with other scatterers, like impurities, phonons, etc.), and it also includes the effect of the electric field (as described above), which makes it time-dependent. The explicit form of this Hamiltonian can be arbitrary for this work, so we do not specify a particular Hamiltonian at this time. The evolution operator of the Hamiltonian with the pump field is denoted $U(t, t_0)$ for the evolution from time t_0 to time t , and it satisfies the equation of motion $i\hbar dU(t, t_0)/dt = \mathcal{H}(t)U(t, t_0)$; it is a unitary operator that also satisfies the semigroup properties $U(t, t) = 1$ and $U(t, t_1)U(t_1, t_0) = U(t, t_0)$. The photocurrent operator J_d represents the current of electrons at the spatial location of the detector (given by \mathbf{R}_d) and with momentum peaked around \mathbf{k}_e . We let $\phi_{\mathbf{k}_e; \mathbf{R}_d}(\mathbf{k})$ be a momentum-space wavefunction peaked about both \mathbf{k}_e and \mathbf{R}_d , and we let $c_{\mathbf{k}_e; \mathbf{R}_d}^\dagger$ be the associated fermionic creation operator to create fermions in the state ϕ . Then, the photocurrent operator satisfies $J_d \approx (\hbar\mathbf{k}_e/m_e)c_{\mathbf{k}_e; \mathbf{R}_d}^\dagger c_{\mathbf{k}_e; \mathbf{R}_d}$, with m_e the mass of the electron. Note that we are suppressing the spin degree of freedom for notational simplicity.

Under the assumption that $\mathcal{H}_{\text{probe}}(t)$ is a weak perturbation to probe the non-equilibrium system, the dominant second-order contribution to the photocurrent becomes

$$\langle \mathbf{J}_d(t) \rangle = \frac{1}{\hbar^2} \int_{t_0}^t dt_1 \int_{t_0}^{t_1} dt_2 \langle U(-\infty, t_2) \mathcal{H}_{\text{probe}}(t_2) U(t_2, t) \mathbf{J}_d U(t, t_1) \mathcal{H}_{\text{probe}}(t_1) U(t_1, -\infty) \rangle, \quad (1)$$

with

$$\langle \mathcal{O} \rangle = \frac{1}{\mathcal{Z}} \sum_n e^{-\beta E_n} \langle n | \mathcal{O} | n \rangle, \quad (2)$$

and $\mathcal{Z} = \sum_n \exp(-\beta E_n)$. Here, E_n is the energy of a many-body eigenstate $|n\rangle$ of the Hamiltonian $\mathcal{H}(-\infty)$ before the pump is turned on, $\beta = 1/k_B T$ is the inverse temperature of the initial equilibrium configuration prior to the pump being turned on, and \mathcal{O} is any operator.

Our material is described as a bulk system with a surface parallel to the x - y plane. We let $\epsilon_{\nu \mathbf{k}_{\parallel}}$ denote the bandstructure with momentum \mathbf{k}_{\parallel} parallel to the surface, and ν denotes all the other quantum numbers (like band index, etc.). The bandstructure is calculated for the initial Hamiltonian $\mathcal{H}(-\infty)$ prior to the pump being turned on. We let $c_{\nu \mathbf{k}_{\parallel}}^{\dagger}$ denote the corresponding electron creation operator; note that we are continuing to suppress spin. The component of $\mathcal{H}(t)$ that corresponds to the absorption of a photon with wavevector \mathbf{q} , frequency $\omega_{\mathbf{q}} = cq$, and creation operator $a_{\mathbf{q}}^{\dagger}$ becomes

$$\sum_{\nu \nu' \mathbf{k}_{\parallel}} s(t) e^{-i\omega_{\mathbf{q}} t} M_{\mathbf{q}}(\nu, \nu', \mathbf{k}_{\parallel}; t) c_{\nu' \mathbf{k}_{\parallel} + \mathbf{q}_{\parallel}}^{\dagger} c_{\nu \mathbf{k}_{\parallel}} a_{\mathbf{q}} \quad (3)$$

with $M_{\mathbf{q}}(\nu, \nu', \mathbf{k}_{\parallel}; t) = \langle \nu' \mathbf{k}'_{\parallel} | (ie\hbar \mathbf{A}_{\text{probe}}(\mathbf{r})/m_e c) \times (\nabla - ie\mathbf{A}_{\text{pump}}(\mathbf{r}, t)/(\hbar c)) | \nu \mathbf{k}_{\parallel} \rangle$ the one-electron matrix element. Here, $\mathbf{A}_{\text{probe}}(\mathbf{r})$ is a product of a spatial envelope function and a plane wave of wavevector \mathbf{q} and $s(t)$ is the probe pulse temporal envelope function. The matrix element itself depends on the vector potential of the pump field, and hence inherits its time dependence. If a tight binding model with a Peierls substitution for the coupling of the system to the vector potential of the pump field is used, the expression for the matrix element M has to be appropriately rewritten.

Introducing the time-reversed low-energy electron diffraction (TRL) states, as described in Reference [18], we then find the total photocurrent for a particular probe pulse is given by

$$\langle \mathbf{J}_d \rangle = \frac{\hbar \mathbf{k}_e}{m_e} \sum_{\nu \nu' \mathbf{k}_{\parallel} \mathbf{k}'_{\parallel}} \phi_{\mathbf{k}_e; \mathbf{R}_d}^*[\mathbf{k}(\nu)] \phi_{\mathbf{k}_e; \mathbf{R}_d}[\mathbf{k}'(\nu')] P_{\mathbf{k}}(s), \quad (4)$$

where $P_{\mathbf{k}}(s)$ is the total probability for a photoelectron to be detected at the detector for a given probe pulse envelope function $s(t)$. We find

$$\begin{aligned} P_{\mathbf{k}}(s) &= \frac{1}{\hbar^2} \sum_{\nu_1 \nu'_1 \mathbf{k}_{\parallel 1}} \sum_{\nu_2 \nu'_2 \mathbf{k}_{\parallel 2}} \int_{-\infty}^{\infty} dt_1 \int_{-\infty}^{\infty} dt_2 M_{\mathbf{q}}^*(\nu_2, \nu'_2, \mathbf{k}_{\parallel 2}; t_2) M_{\mathbf{q}}(\nu_1, \nu'_1, \mathbf{k}_{\parallel 1}; t_1) s(t_2) s(t_1) e^{i\omega_{\mathbf{q}}(t_2 - t_1)} \\ &\times \langle c_{\nu_2 \mathbf{k}_{\parallel 2}}^{\dagger}(t_2) c_{\nu'_2 \mathbf{k}_{\parallel 2} + \mathbf{q}_{\parallel}}(t_2) c_{\nu'_1 \mathbf{k}'_{\parallel 1}}^{\dagger}(t_1) c_{\nu_1 \mathbf{k}_{\parallel 1}}(t_1) c_{\nu'_1 \mathbf{k}_{\parallel 1} + \mathbf{q}_{\parallel}}^{\dagger}(t_1) c_{\nu_1 \mathbf{k}_{\parallel 1}}(t_1) \rangle. \end{aligned} \quad (5)$$

All fermionic creation and annihilation operators are in the Heisenberg representation with respect to the Hamiltonian with the pump present; i.e., $\mathcal{H}(t)$.

The expectation value is a three-particle non-equilibrium many-body Green's function, which is challenging to evaluate. However, it is a good approximation to assume that the high energy electrons (for example, the TRL state electrons indexed by ν and ν') do not strongly interact with other electrons, and that the pump pulse does not significantly alter them (because they reside primarily outside the region where the pump pulse lives). In this case, they can be treated as noninteracting band-like

electrons, which allows the three-particle Green’s function to be factorized, just like in Wick’s theorem, which yields

$$\begin{aligned} & \langle c_{v_2\mathbf{k}_{\parallel 2}}^\dagger(t_2)c_{v_1\mathbf{k}_{\parallel 1}}(t_1) \rangle_{\mathcal{H}} \times \langle c_{v'_2\mathbf{k}_{\parallel 2}+\mathbf{q}_{\parallel}}(t_2)c_{v'\mathbf{k}'_{\parallel}}^\dagger(t) \rangle_{\mathcal{H}_0} \times \langle c_{v\mathbf{k}_{\parallel}}(t)c_{v'_1\mathbf{k}_{\parallel 1}+\mathbf{q}_{\parallel}}^\dagger(t_1) \rangle_{\mathcal{H}_0} \\ & \simeq \langle c_{v_2\mathbf{k}_{\parallel 2}}^\dagger(t_2)c_{v_1\mathbf{k}_{\parallel 1}}(t_1) \rangle_{\mathcal{H}} \delta_{v_2,v'}\delta_{v'_1,v}\delta_{\mathbf{k}_{\parallel 2}+\mathbf{q}_{\parallel},\mathbf{k}'_{\parallel}}\delta_{\mathbf{k}_{\parallel 1}+\mathbf{q}_{\parallel},\mathbf{k}_{\parallel}} e^{i[(\epsilon_{v'\mathbf{k}'_{\parallel}}-\mu)(t-t_2)-(\epsilon_{v\mathbf{k}_{\parallel}}-\mu)(t-t_1)]} \end{aligned} \quad (6)$$

Here, the expectation value involving the low-energy electrons is evaluated as a single-particle non-equilibrium lesser Green’s function evolving according to the full Hamiltonian \mathcal{H} . The high-energy averages are non-interacting equilibrium single-particle Green’s functions evaluated with respect to the band Hamiltonian \mathcal{H}_0 , which includes no pump and no interactions of the electrons (with themselves or anything else). These high energy averages can then be explicitly evaluated, as shown in the last line above, where we have further assumed that the initial temperature is small compared to the energies of the photo-emitted electrons and the pump does not excite the electrons to too high an energy.

A further approximation is to assume that the momentum distribution of the electrons arriving at the detector is sharp. Then, we have $\mathbf{k}_e = \mathbf{k}(v) = \mathbf{k}'(v')$ and $v = v' = v_e$. Finally, we also have $\epsilon_{v_e\mathbf{k}_{e\parallel}} - \mu = \hbar\omega_{\mathbf{q}} - \hbar\omega$, with $\hbar\omega$ the excitation energy remaining in the system after the photoemission process. This then yields the main result for the TR-ARPES formalism

$$\begin{aligned} P_{\mathbf{k}}(s) & \simeq -\frac{i}{\hbar^2} \sum_{v_1,v_2} \int_{-\infty}^{\infty} dt_1 \int_{-\infty}^{\infty} dt_2 M_{\mathbf{q}=0}^*(v_2, v_e; \mathbf{k}_{e\parallel}; t_2) M_{\mathbf{q}=0}(v_1, v_e; \mathbf{k}_{e\parallel}; t_1) s(t_2) s(t_1) e^{i\omega(t_2-t_1)} \\ & \times G_{v_1\mathbf{k}_{e\parallel},v_2\mathbf{k}_{e\parallel}}^<(t_1, t_2), \end{aligned} \quad (7)$$

where we assumed the photon wavevector is much smaller than the electron wavevectors. The two-time non-equilibrium lesser Green’s function is

$$G_{v_1\mathbf{k}_{e\parallel},v_2\mathbf{k}_{e\parallel}}^<(t_1, t_2) = i\langle c_{v_2\mathbf{k}_{e\parallel}}^\dagger(t_2)c_{v_1\mathbf{k}_{e\parallel}}(t_1) \rangle_{\mathcal{H}} \quad (8)$$

and the operator averages are taken with respect to the full Hamiltonian. We label the photoresponse with the symbol s , since the envelope function of the probe Hamiltonian encodes the time delay with respect to the pulse. Since the integrals over time extend from $-\infty$ to ∞ , we count *all* photoelectrons which reach the detector from the applied pulses.

2.2. Gauge Invariance of the TR-ARPES Signal

In the previous subsection, we calculated the TR-ARPES signal in a specific gauge (the so-called Hamiltonian gauge where the vector potential is nonzero, but the scalar potential vanishes). If the time delay between the pump and the probe is long enough that the pump and the probe pulses do not overlap in time, then the vector potential vanishes, and the results are manifestly gauge invariant. In addition, if one calculates the *total* photoemission response, integrating over momentum, the final result is also manifestly gauge invariant, even when the pump and probe pulses do overlap. However, in this latter case, if one is interested in the *angle-resolved* response, then, while the full expression in Equation (7) is easily shown to be gauge-invariant, one must exercise care with the approximations one inevitably makes—especially the approximation of ignoring the momentum dependence of the matrix elements (which we will refer to as the constant matrix element approximation)—in evaluating the response function to ensure that it continues to be gauge invariant. To date, no one has properly formulated such gauge-invariant prescriptions when the effects of a surface, of a three dimensional dispersing band structure, or of the pump pulse on the propagation of the photoelectrons are important. We will not solve this problem here either. Instead, we focus on the simplest scenario, where (in addition to the constant matrix element approximation) the spatial dependence of the electric field is neglected (which is an excellent approximation for optical or

infrared pump pulses) and where the Green's function depends only on the momenta parallel to the sample surface.

In this case, one possible prescription is to follow Bertoncini and Jauho [20] and replace the lesser Green's function by its gauge invariant modification:

$$G_{\nu_1 \mathbf{k}_{e\parallel}, \nu_2 \mathbf{k}_{e\parallel}}^<(t_1, t_2) \rightarrow \tilde{G}_{\nu_1 \mathbf{k}_{e\parallel}, \nu_2 \mathbf{k}_{e\parallel}}^<(t_1, t_2) \equiv G_{\nu_1 \bar{\mathbf{k}}_{e\parallel}, \nu_2 \bar{\mathbf{k}}_{e\parallel}}^<(t_1, t_2), \quad (9)$$

with

$$\bar{\mathbf{k}}_{e\parallel}(t_1, t_2) = \mathbf{k}_{e\parallel} - \frac{e}{\hbar c} \frac{1}{t_1 - t_2} \int_{-(t_1-t_2)/2}^{(t_1-t_2)/2} dt' \mathbf{A}_{pump} \left(\frac{t_1 + t_2}{2} + t' \right). \quad (10)$$

While it is straightforward to show that the TR-ARPES signal is nonnegative in a gauge, it remains challenging to show non-negativity for the gauge-invariant signal; physically, however, this result must always be true.

If the TR-ARPES signal is integrated over angles to give the total TR-PES signal, then the constant matrix element approximation will always yield a gauge-invariant result. This is because quantities evaluated in a gauge and summed over all momentum are manifestly gauge-invariant. This procedure has been employed for multiband models of charge-density-wave insulators [23] and topological insulators [24].

2.3. Constant Matrix-Element Approximation

For simplicity, we focus on quasi-2D materials, where $\mathbf{k}_{\parallel} = \mathbf{k}$ is the 2D wavevector. This approximation works well for many interesting systems including high T_c superconductors, graphene, transition-metal dicalchogenides, and many topological insulators. If only one band (ν_1) contributes at low energy, then the constant matrix element approximation becomes trivial. Namely, the gauge-invariant TR-ARPES signal becomes

$$P_{\mathbf{k}}^{GI}(s) \simeq -\frac{i}{\hbar^2} |M|^2 \int_{-\infty}^{\infty} dt_1 \int_{-\infty}^{\infty} dt_2 s(t_2) s(t_1) e^{i\omega(t_2-t_1)} G_{\nu_1 \bar{\mathbf{k}}_e(t_1, t_2), \nu_1 \bar{\mathbf{k}}_e(t_1, t_2)}^<(t_1, t_2), \quad (11)$$

and there is no freedom in the approximation (we dropped the parallel symbol since the wavevector is in 2D only here). As mentioned before, the result in a gauge is non-negative as a trivial application of Bochner's theorem [19,25], but for the gauge-invariant result, it is difficult to prove non-negativity.

For higher numbers of bands, the matrix elements—even when assumed to be independent of momentum—can have different values for different bands. By examining the structure of the photoemission response, we see that it involves a product of a vector (of the different matrix elements) times the lesser Green's function matrix times the Hermitian conjugate of the vector of matrix elements, which is a quadratic form. Such an object is nonnegative if all eigenvalues of the matrix are nonnegative. However, more important than that is the fact that the result requires a summation over *all matrix elements of $G^<$* . Hence, it is not a matrix invariant purely of the $G^<$ matrix, and if one changes the basis—especially if one changes the basis as a function of time—then the matrix elements must also be changed according to the unitary transformation that effects the change of basis, and hence will become functions of time. This is true even if we choose the matrix elements to be the same for all the bands. Of course, the total photoemission signal is independent of whatever basis is chosen to represent the Green's function matrix. The vector of matrix elements changes its values as one changes the basis, so that the final result of the vector–matrix–vector product does indeed remain invariant of the basis. However, one cannot assume that the individual matrix elements are constants independent of the basis chosen for the bands. They can change as the basis changes, and indeed must change for the final result to be invariant. Emphasizing this subtle point is the main message of this work.

These results imply that the constant matrix element approximation becomes more complicated when multiple bands are involved in the low-energy dynamics of the material. Before we discuss the implications of these choices further, we first want to discuss the situation if we consider the

angle-integrated photoemission spectroscopy. In the constant matrix element approximation, we neglect the \mathbf{k}_e dependence of the matrix element, so the TR-PES signal becomes

$$P_{\text{local}}^{GI}(s) \simeq -\frac{i}{\hbar^2} \int_{-\infty}^{\infty} dt_1 \int_{-\infty}^{\infty} dt_2 s(t_2) s(t_1) e^{i\omega(t_2-t_1)} \sum_{\nu_1 \nu_2} M_{\nu_1} G_{\nu_1, \nu_2}^{<\text{local}}(t_1, t_2) M_{\nu_2}^* . \quad (12)$$

It is common to perform the calculations in a basis that is diagonal in band space. In this case, if we also choose all of the matrix elements to be the same, then the TR-PES signal is given by the trace of the Green's function, which can give the false impression that the result is always the trace of the Green's function matrix. However, as we discussed above, this is not the case.

Sometimes it might be convenient to work in the instantaneous diagonal basis for a given wavevector. Because the Peierls substitution shifts the wavevector in a time-dependent fashion, the basis changes with time (for example, the Bloch wave functions of the instantaneous one electron component of $\mathcal{H}(t)$ would have this character). In this circumstance, one can choose to make a further approximation of choosing the matrix elements to be constant (and thereby factor out of the calculation)—even though the basis is being changed for each time point—and use the trace formula, since the Green's function is diagonal in this basis. It should be noted that doing so introduces an uncontrolled approximation into the system, and could result in a photoemission spectra which became negative. Nevertheless, it may be an interesting approximation to make if no further information is known about the matrix elements [26,27].

The general conclusion that we make for the constant matrix element approximation is that it can become problematic when it is applied to systems with more than one band. The best way to proceed (if feasible) is to determine a reasonable guess for the ratios of the matrix elements in the different bands, keep track of all basis-changing operations during the calculation, transform the matrix elements accordingly, and be sure to evaluate the final result as the product of the Green's function matrix with the matrix element vector and its Hermitian conjugate. In this case, we expect that the TR-ARPES signal will be the most physical result and will maintain non-negativity. If it becomes negative, this would then be a fault of the constant matrix element approximation, which would need to be fixed to correct the error.

The constant matrix element approximation is much more robust for the case of angle-integrated spectra. In this situation, the final formula in Equation (12) holds, one can assume the matrix elements are the same for each band, and if the local Green's function is diagonal in the band basis, then the signal is proportional to the trace of the local lesser Green's function.

3. Discussion and Conclusions

In this work, we helped clarify a number of subtle points associated with TR-ARPES studies when the number of low-energy bands is larger than one. These effects become particularly important for materials with a basis like graphene, or most topological insulators. Our focus was on how one generalizes the constant matrix element approximation from one to many bands. It turns out that while the single-band case is completely determined with no extra freedom, the case with multiple bands is more complex and has extra degrees of freedom associated with them.

In particular, there can be a different matrix element associated with each band; but more than that, if the basis for the Green's function is changed, then one is forced into considering matrix elements that are no longer constant, since the new basis relates to the old by a unitary transformation which may change with time. We also discussed that in the case when the Green's function is diagonal, then the constant matrix element approximation is proportional to the trace of the Green's function if all matrix elements are chosen to be the same. Finally, we described how one can make a further uncontrolled approximation of using the trace formula with the same constant matrix elements for different bases, but in that case, it is likely that the spectra will become negative for some times and frequencies.

We ended by discussing the case for the constant matrix element approximation when one considers angle-integrated spectra. Here, there are far fewer degrees of freedom in the results, and they are also manifestly gauge-invariant.

In the future, it would be interesting to discuss both the constant matrix element approximation and the issue of gauge invariance within a fully developed theory starting from first principles, rather than in an ad hoc fashion, as we currently do. It also would be useful and interesting to understand what functional dependence of the matrix elements is most critical for understanding real spectra. Is it the wavevector dependence or the band index dependence? How do these results affect the final TR-ARPES spectra?

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Abbreviations

The following abbreviations are used in this manuscript:

TR-ARPES	time-resolved angle-resolved photoemission spectroscopy
TR-PES	time-resolved photoemission spectroscopy
LEED	Low-energy electron diffraction
TRL	time-reversed LEED

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