Erratum: Theoretical Description of time-Resolved Photoemission Spectroscopy: Application to Pump-Probe Experiments [Phys. Rev. Lett. 102, 136401 (2009)]

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We discovered that some of the equations and discussions presented in our Letter have the limitation that they are not general enough to ensure manifest gauge invariance in contexts where the pump pulse continues to be on when the probe pulse is operative, and can hence lead to erroneous results. These limitations are easily corrected. Unfortunately, due to space constraints for an erratum in Physical Review Letters, the corrected formulas are to be found elsewhere. We will carefully show where in the discussion below.

To begin with, Eq. (3) is not general enough. The probe Hamiltonian $\mathcal{H}_{\text{probe}}(t_1)$, must have additional time dependence due to the vector potential of the pump pulse. Hence, Eq. (3) is correctly rewritten with $M_{\mathbf{q}}(\nu, \nu'; \mathbf{k}_{\parallel}) \rightarrow M_{\mathbf{q}}(\nu, \nu'; \mathbf{k}_{\parallel}; t_1)$, given by the *modified* expression $\langle \nu' \mathbf{k}'_{\parallel} | (ie\hbar \mathbf{A}_{\text{probe}}(\mathbf{r})/m_e c) \cdot (\nabla - ie\mathbf{A}_{\text{pump}}(\mathbf{r}, t_1)/(\hbar c)) | \nu \mathbf{k}_{\parallel} \rangle$, which 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 updated. Then, the expression for $P(t)$ in Eq. (4) is modified by adding t dependence to the matrix elements and moving them into the integrals with t_1 being the argument for the matrix element with the 1 subscripts and similarly for t_2 . Furthermore, in the context of the approximations leading to the factorization in Eq. (6), and those discussed in the paragraph following Eq. (6), Eq. (6) must be modified too. The modified equation can be found as Eq. (5) of Ref. [1]. In addition, if the pump pulse is on when the probe pulse ejects the photoelectron, there is a further approximation involved in going from the lhs to the rhs of Eq. (6), in that the effect of the pump-pulse on the propagation of the photoexcited electron has been neglected. Given this approximation, Eq. (7) for $P(t)$ has to be modified with the two matrix elements now going under the integrand and with the same time arguments t_1 and t_2 . Please see Eqs. (6) and (7) of Ref. [1] for the corrections. We note that in contexts where the pump pulse is turned off before the probe pulse is turned on, the matrix elements M no longer have any time dependence, and all of the new expressions reduce to the ones given in the original letter.

It is customary in calculations of PES of strongly correlated layered materials not to calculate the matrix elements, but instead to replace them by constants and furthermore to ignore the indices ν_1 and ν_2 in Eq. (8). But, then the nonequilibrium lesser Green's function in Eq. (8) calculated in the presence of the pump field becomes gauge dependent. To fix this, we follow Bertoncini and Jauho [2], and replace the lesser Green's function by its gauge invariant modification as shown in the discussion around Eq. (10) of Ref. [1]. It remains a theoretical challenge to *derive* the appropriate gauge independent prescription, in the contexts where the presence of the surface, three-dimensionally dispersing band structure effects, the effect of the pump pulse on the propagation of the photoelectrons, etc., are important.

In addition, we must emphasize that we have kept only the *dominant* contribution to the detected current to second order in $\mathcal{H}_{\text{probe}}$, and not the *entire* contribution. There are two other contributions to $\langle \mathbf{J}_d \rangle(t)$ that are formally of order $(\mathcal{H}_{\text{probe}})^2$ which in principle need to be included, which yield two additional terms in the expression for $P(t)$. The full formulas can be found in Sec. 5 of Ref. [1]. The physical processes corresponding to the extra terms require the detected electron to come right out of the (*time-evolved*) *initial state, before the photon is absorbed*, which can happen only when that state has an electron excited to a high enough band (TRL) state that it will come out of the sample. When the pumped system is describable as thermalized with an effective electron temperature T_e , the Boltzmann probability for this is proportional to $\exp[-\epsilon_p/(k_B T_e)]$ where $\epsilon_p = \epsilon_{\nu \mathbf{k}_{\parallel}} - \mu$ is the (excitation) energy of the detected electron measured from the chemical potential of the system. This is clearly small as long as ϵ_p is much larger than $(k_B T_e)$ and can happen even if the kinetic energy of the detected electrons is not very large, e.g., if ϵ_p is only slightly larger than the work function, but the latter is much larger than $(k_B T_e)$. Even when the pumped system is not in a thermal distribution, electrons can only be excited to such higher bands either via a tunneling process, or via multiphoton absorption processes; in both cases one expects the population that is excited and the contributions from the neglected terms, to be small. But there might be special experimental circumstances where the extra terms are measurable, and need to be taken into account.

Finally, there is an issue about whether one can use a constant matrix element approximation for multiband systems. It turns out that one cannot, and a full explanation for why is given in Ref. [3].

- [1] J. K. Freericks, H. R. Krishnamurthy, M. A. Sentef, and T. P. Devereaux, *Phys. Scr.* **2015**, 014012 (2015).
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