Comment on “Free-Electron–Bound-Electron Resonant Interaction”

The interaction between a beam electron and a single optical excitation in a sampled structure is generally weak, thus making it difficult to study atoms and molecules with electron microscopes. To remedy this situation, Gover and Yariv [1] propose to prepare the electron wave function in a state consisting of a periodic train of tightly focused pulses, such as those demonstrated using photon-induced near-field electron microscopy (PINEM) [2,3] or stimulated Compton scattering [4]. The authors focus on a two-level sample excitation (2LSE) and describe its interaction with the beam through the electric field generated by a classical current that has the same space-time profile as the probability density associated with the electron wave function. The purpose of this Comment is to show that such treatment of the electron wave as a classical current distribution leads to conclusions that radically differ from those obtained by a quantum description of the electron.

The argumentation in Ref. [1] is as follows: when assimilating the incident electron to a time-periodic classical current, its interaction with the 2LSE can be described through an effective external potential that contains harmonics of the fundamental PINEM sideband frequency $\omega_p$ and can resonantly excite the sample if the 2LSE frequency difference is a multiple of $\omega_p$. Even for weak electron-sample interaction, $N$ electrons can then produce an excitation probability $\propto N^2$ by accumulating quantum kicks from individual electrons, regardless of their relative arrival times, as long as their temporal separation is small compared with the excitation lifetime. We show next that these conclusions are not maintained when the electrons are described quantum mechanically rather than classically.

In a general quantum description of the system formed by the beam electron and the sample, we expand the total wave function $|\psi(t)\rangle = \sum_{k} \alpha_{kn}(t) e^{-i(\epsilon_{kn} + q\omega_0)t}|kn\rangle$ in a complete basis of electron states $|k\rangle$ (momentum $hk$, energy $\epsilon_{kn}$, and wave function $e^{ikz-ix_0t}$ along the propagation direction $z$) and sample states $|n\rangle$ (energy $\hbar \omega_{an}$). Elements of this basis set are eigenstates of the noninteracting Hamiltonian: $\hat{H}_0|kn\rangle = h(\epsilon_{k} + \omega_{an})|kn\rangle$. Now, we consider an electron-sample interaction Hamiltonian $\hat{H}_I$ and insert the above expression for $|\psi(t)\rangle$ into the Schrödinger equation $(\hat{H}_0 + \hat{H}_I)|\psi\rangle = i\hbar \partial_t|\psi\rangle$. The expansion coefficients $\alpha_{kn}$ are thus found to satisfy the equation $i\hbar \alpha_{kn} = \sum_{k'\neq k} e^{i(\epsilon_{k'}+q\omega_0)t} \langle k|\frac{\partial}{\partial t} H_0|k'\rangle \alpha_{k'n}$, where $\epsilon_{k'} = \epsilon_k - q\omega_0$ and $\alpha_{k'n} = \alpha_{kn} - \alpha_{kn'}$. At this point, we make two well-justified assumptions [5] also adopted in Ref. [1]: (1) the electron-sample interaction is weak enough to treat it to first order, and (2) the electron momentum remains tightly focused around a value $\hbar k_0$, so only $\alpha_{kn}$'s with $|k - k_0| \ll k_0$ contribute.

Assumption (1) allows us to retain only the $n' = 0$ term (initial sample state), leading to the equation of motion $\hbar \alpha_{kn} = \sum_{k'} e^{i(\epsilon_{k'} + q\omega_0)t} \langle kn|\frac{\partial}{\partial t} H_0|k'\rangle \alpha_{k'0}$, where $\alpha_{k'0} = \alpha_{k0}(t = -\infty)$ defines a general incident electron wave function. Direct integration yields $\alpha_{kn}(t = \infty) = (-2\pi/\hbar) \sum_{k'} \delta(\epsilon_{k'} + q\omega_0) \langle kn|\frac{\partial}{\partial t} H_0|k'0\rangle \alpha_{k'0}$ after interaction.

Assumption (2) implies that terms inside the $k'$ sum depend on $k'$ only through the difference $q = k' - k$ because the electron wave functions contribute with $e^{iqz}$ inside the interaction matrix element $V_{kn} \equiv \langle kn|\frac{\partial}{\partial t} H_0|k'0\rangle$, and furthermore, we can approximate $\epsilon_{k'} \approx -q\nu$, where $\nu = \partial_{k}\epsilon_{k}|_{k = k_0}$ is the electron velocity. This allows us to write the probability of exciting the sample to state $n$ as

$$P_n = \sum_k |\alpha_{kn}(\infty)|^2 = \frac{4\pi^2}{k_0^3} \sum_k \delta(q\nu - \omega_0) V_{kn}^2,$$

where the summation variable $k'$ is changed to $q$ and we have used the normalization condition $\sum_k |\alpha_{kn}|^2 = 1$ for the incident electron. We conclude that the excitation probability $P_n$ [Eq. (1)] is independent of the electron wave function profile under the assumptions of first-order interaction and nonrecoil, provided the sample is initially prepared in an eigenstate. Incidentally, retardation effects are included in this derivation for a proper choice of the Hamiltonian $\hat{H}_1$. The excited states can also describe photon emission by interaction with gratings or other elements (i.e., the present fully quantum analysis applies to any choice of sample system and interaction Hamiltonian $\hat{H}_1$).

The results from Ref. [1] are therefore not maintained when the electron is described quantum mechanically, rather than treated as a classical current distribution. In particular, no resonant effects associated with multiples of the PINEM sideband spacing $\omega_p$ are expected. Additionally, a straightforward extension of the above quantum formalism to $N$ electrons readily reveals a contribution $\propto e^{-i\omega_0t}$ to the excitation amplitude by each electron $j$, $t_j$ refers to the centroid of the electron wave function; the excitation probability is thus $\propto N + \sum_j \sum_{j \neq j} e^{i\omega_0(t_j-t_j')} at most, where the second term vanishes when averaging over random $t_j$'s spanning a large interval compared with the excitation optical cycle, even if electrons share a nearly identical wave function (apart from translations in time by $t_j$) and their relative delays $|t_j - t_j'|$ are less than the lifetime of the sampled excitation [7]. In conclusion, an $N^2$ contribution is not expected, although superradiance is of course trivially recovered if $|t_j - t_j'| \ll 2\pi/\omega_0$ [8].

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The last paragraph of the Comment contains information that is misleading in the context of the commented paper: while the former shows that the excitation probability is independent of the wave function for individual electrons (i.e., no resonant effects are found from a quantum mechanical treatment of the electron even if it is PINEM-modulated), the extension to $N$ electrons discussed in the noted paragraph assumes that they are prepared with the same wave function profile, but random arrival times. For $N$ PINEM-modulated electrons, this condition is not met because their wave functions contain a temporal modulation factor that is the same in all of them (as imposed by the modulating laser), leading to an excitation probability scaling as $N^2$ (see Ref. [7] in the Comment for a first-principles derivation). In conclusion, while the classical treatment presented in the commented paper for an individual electron leads to different results from the quantum description offered in the Comment, an extension of the latter to $N$ PINEM-modulated electrons produces an $N^2$ scaling of the excitation probability, in agreement with the prediction in the commented paper based on a description of the electrons as classical charge distributions.