

Emergence of electron coherence and two-color all-optical switching in MoS₂ based on spatial self-phase modulation

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Generating electron coherence in quantum materials is essential in optimal control of many-body interactions and correlations. In a multidomain system this signifies nonlocal coherence and emergence of collective phenomena, particularly in layered 2D quantum materials possessing novel electronic structures and high carrier mobilities. Here we report nonlocal ac electron coherence induced in dispersed MoS₂ flake domains, using coherent spatial self-phase modulation (SSPM). The gap-dependent nonlinear dielectric susceptibility $\chi^{(3)}$ measured is surprisingly large, where direct interband transition and two-photon SSPM are responsible for excitations above and below the bandgap, respectively. A wind-chime model is proposed to account for the emergence of the ac electron coherence. Furthermore, all-optical switching is achieved based on SSPM, especially with two-color intraband coherence, demonstrating that electron coherence generation is a ubiquitous property of layered quantum materials.

electron coherence | transition metal dichalcogenide | self-phase modulation | optical switching | emergent phenomena

Recently 2D layered quantum materials have attracted tremendous interest since the discovery of graphene a decade ago (1). Various layered materials, ranging from boron nitride sheets to transition metal dichalcogenides and from topological insulators to high-temperature superconductors, have been intensively investigated (2–11). Strict 2D atomic crystals can now be produced at a macroscopic scale, using a variety of methods (11, 12). Among them molybdenum disulfide (MoS₂) and related layered quantum materials are particularly interesting due to their novel optical properties and potential valleytronics applications (4, 5, 8, 9) at a thickness of monolayer and few layers (2, 10, 13). Layered materials share common physical properties rooted in their ubiquitous 2D quantum nature, for which achieving pure coherence among electrons (lattices) is of particular interest (14–19). The presence of multiple domains is ubiquitous in many known 2D quantum materials, ranging from stripe-order cuprate superconductors to polycrystalline strongly correlated systems. For example, phase locking between different layers of stripe orders is crucial for enhancing the superconducting phase in layered high-temperature superconductors (20, 21).

In this work we demonstrate unambiguously that nonlocal and intraband ac electron coherence, of which the electronic wave function oscillates at an optical frequency of 10^{14} Hz, can be generated in separate MoS₂ flakes, using spatial self-phase modulation (SSPM). The SSPM is a coherent third-order nonlinear optical process systematically investigated decades ago (22), where the nonlinear optical susceptibility $\chi^{(3)}$ is uniquely determined by the laser-intensity-dependent refractive index $n = n_0 + n_2 I$, where n_0 and n_2 are linear and nonlinear refractive indexes, respectively. If this effect is strong enough in a material, the phenomenon of self-focusing can be directly observed. The SSPM is also frequently referred to as the optical Kerr effect or the ac Kerr effect (note the difference from the regular Kerr effect). It is parallel to the other

third-order nonlinear optical processes, such as third harmonic generation (THG) and four-wave mixing (FWM).

Our investigations show that gap-dependent SSPM is a general method for inducing electron coherence in 2D layered materials. Monolayer and few-layer MoS₂ (and other similar layered materials) have finite bandgaps, which lift the obstacle to versatile electronic and optical applications (2, 23, 24). Because SSPM using below-gap photons has not been reported, MoS₂ provides an ideal test platform for observing SSPM in finite-bandgap materials. A model has been developed to account for the coherence emergence process and theoretical calculations have been carried out to unravel the underlying mechanism. Moreover, we demonstrate all-optical switching based on SSPM, which employs intraband electron coherence. This optical switch has multiple advantages, including weak-control-strong performance, cascade-possible, and high-contrast two-color switching.

Results and Discussion

Fig. 1 displays the experimental setup and typical SSPM patterns obtained from the flake domains of MoS₂ in a suspension (*Methods*). It can be seen from Fig. 1*B* that the sample is mainly composed of MoS₂ flakes with a thickness of 30 ~ 55 layers. In Fig. 1 *D–F* we show the SSPM patterns obtained using 400-nm ultrafast laser pulses, 532-nm continuous wave (cw) laser beams, and 800-nm ultrafast laser pulses, respectively. The SSPM is a nonlinear optical process in that the phase (rather than the intensity) of the output beam depends on the input intensity (*Methods*).

Significance

Generating electron coherence is nontrivial in that most sophisticated electronic experimental methods are incoherent or cannot be used to induce and detect collective states. By using coherent spatial self-phase modulation (SSPM) (a nonlinear optical property) we observed the emergence of electron coherence in a gapped quantum material, MoS₂. By observing gap-dependent SSPM we discovered that it is a ubiquitous property of two-dimensional layered quantum materials. Furthermore, we demonstrate that this ac electron coherence can be harnessed to realize two-color all-optical switching with superb performance.

Author contributions: J.Z. designed research; Y.W., Q.W., F.S., and J.Z. performed research; C.C. and S.M. obtained theoretical absorption; Q.W. and J.Z. derived wind chime model; Y.W., S.M., and J.Z. analyzed data; and S.M. and J.Z. wrote the paper.

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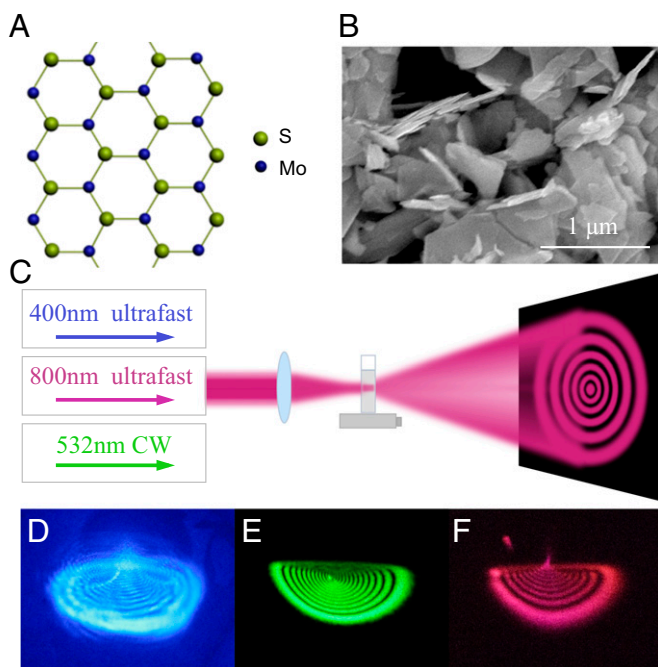


Fig. 1. (A) Schematic lattice structure of MoS₂. (B) Scanning electron microscopy image of the MoS₂ flakes. (C) Schematic experimental setup. (D–F) The SSPM patterns generated using 400-nm, 532-nm, and 800-nm laser beams, respectively.

To unambiguously verify that we have observed SSPM, we performed intensity-dependence measurements. Fig. 2 shows that the number of rings N increases nearly linearly with the laser intensity, until the saturation is reached. By this linear dependence the optical nonlinear coefficient n_2 , hence $\chi^{(3)}$, can be obtained directly (Fig. 2 and *Methods*). Because nearly all of the light is diffracted off the incidence direction and the Rayleigh scattering is low, ideally it is a purely coherent third-order nonlinear optical process. This pure coherence must originate from the coherence within the sample—the electron coherence, where the electronic wave function preserves a definite phase (see discussion below). In the case that photoexcited carriers become out of phase due to collisions, impurity scatterings, and boundary reflections, they reassume their coherence transiently (at a time-scale of femtoseconds) under the external light pulse.

Next we consider nonlocal electron coherence. In our sample, each flake is a domain. Charge carriers in any form (including photocarriers, excitons, free electron–hole pairs, and intrinsic electrons and holes) in different and the same domains are initially completely out of phase. Besides, each domain has an arbitrary orientation. Upon light interaction, photoinduced quasiparticles interact with the electric field (Fig. 3A) and assume its local phases.

We propose a wind-chime model to describe the emergence of electron coherence from the nonlocal domains driven by SSPM (Fig. 3A). Initially there exists an arbitrary orientation angle θ between a MoS₂ flake (thus the electrical polarization it contains) and the electric field. Due to energy relaxation, the electric field reorients the flakes so that each domain contains an axis parallel to the polarization of the external field, as if each domain is “hung” by a vertical “thread.” This image mimics that of a wind chime (Fig. 3A). Once the wind chime is formed, the electron coherence is completely set up within and among each of the different domains. This scenario depicts the emergence of nonlocal electron coherence as a complex collective behavior.

We first show that the model is reasonable. Assuming a circular disk shape for the MoS₂ flake domains, we derived (*SI Text*,

section S1) that the rotation torque generated by the laser pulse $\mathcal{M} = \int |\mathbf{P} \times \mathbf{E}| dV$ has a magnitude of

$$\mathcal{M} = \frac{1}{4} \sin 2\theta \frac{(\epsilon_r - 1)}{\epsilon_r} \epsilon_0 E_0^2 \pi R^2 h e^{-(2(ct-z)^2/c^2\tau^2)}, \quad [1]$$

where R is the disk radius, h is the disk thickness, and τ is the pulse width. Taking the Newton fluid with constant viscosity coefficient η and linear velocity v at the interface, the rotation torque \mathcal{V} due to the viscous force is

$$\mathcal{V} = \eta \int_0^\pi \frac{dv}{dr} (R \sin \varphi) (\xi \cdot 2\pi R \sin \varphi) (R d\varphi) = \pi \eta \Omega \xi R^3, \quad [2]$$

where Ω is the rotation velocity and ξ is the portion of the fluid globe that is rotating together with the disk. Hence the rotation angle accumulated due to each single light pulse is (*SI Text*, *section S1*)

$$\delta\theta = \int_0^T \Omega dt \approx \int_0^\infty \Omega_0 e^{-(\pi \eta \xi R^3 / (J_{\text{MoS}_2} + J_{\text{Solution}})) t} dt = \frac{(\epsilon_r - 1)}{\epsilon_r} \frac{2T I h}{4\eta \xi R c} \sin 2\theta, \quad [3]$$

where T is the repetition period. The time needed for the pattern formation is (*SI Text*, *section S1*)

$$T = \frac{\epsilon_r \pi \eta \xi R c}{1.72(\epsilon_r - 1) I h} = 0.3 \text{ s}, \quad [4]$$

where we have used the values $\epsilon_r = 3.33$ for MoS₂, $\eta = 3.2 \times 10^{-4}$ Pa·s for acetone, $R \sim 1 \mu\text{m}$, $h \sim 10 \text{ nm}$, $I = 250 \text{ W/cm}^2$, and $\xi \sim 0.03$ in our experiment. As a result, T does not depend on T . Thus, for both pulsed and cw excitations the results are identical, which has been verified in our experiment. To compare with the experiment, we recorded the formation processes of the ring patterns (see snapshots in Fig. 3B and C). The ring number and diameter both increase monotonously with time until the maximum is reached after 0.20 s. This compares well with the predicted value by the wind-chime model. Thus, the observed

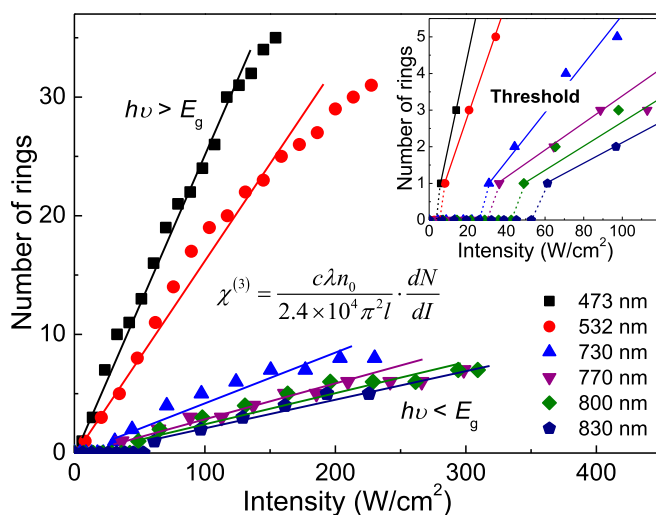


Fig. 2. Intensity dependence of ring numbers and bandgap dependence of $\chi^{(3)}$. The slopes directly illustrate the nonlinear coefficients $\chi^{(3)}$. The solid lines are guides to the eye. (Inset) The thresholds to observe the SSPM patterns at different photon energies.

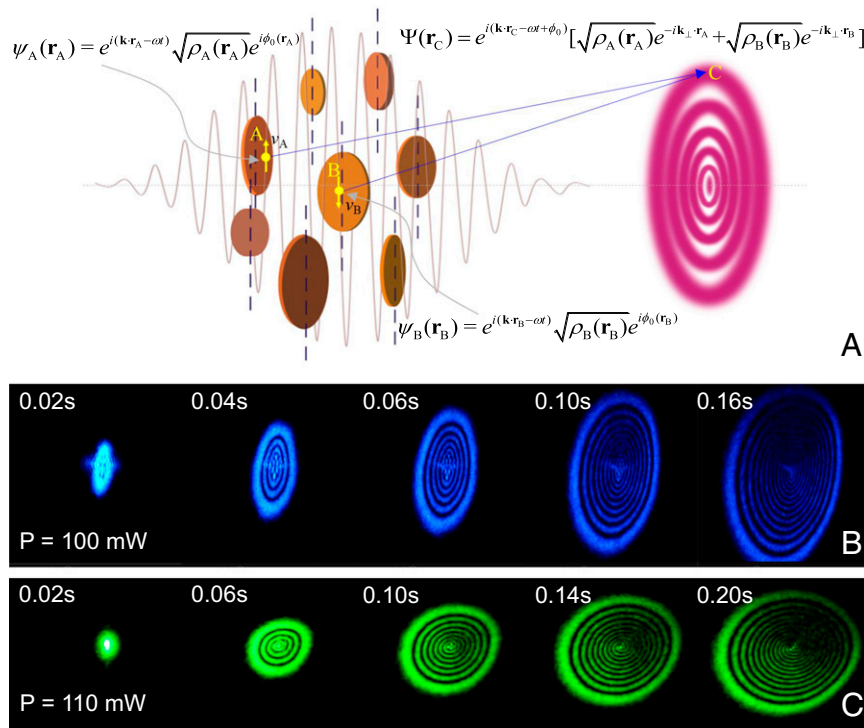


Fig. 3. Wind-chime model and the emergence of electron coherence. (A, Left) Collective response of the MoS₂ flakes to the laser beam electric field, being aligned to form a “wind chime.” (A, Right) SSPM pattern formed. (B and C) Snapshots of the pattern formation at 473-nm and 532-nm laser beam excitations, respectively. The whole process takes ~ 0.20 s, which is consistent with the wind-chime model prediction (main text).

time for the ring formation is a strong experimental support to the wind-chime model.

We then discuss the emergence of the ac electron coherence. Writing the electronic wave function at \mathbf{r}_A as $\psi_A(\mathbf{r}_A) = \sqrt{\rho_A(\mathbf{r}_A)} e^{i\phi(\mathbf{r}_A)}$, with $\psi_A^*(\mathbf{r}_A) \cdot \psi_A(\mathbf{r}_A) = \rho_A(\mathbf{r}_A)$ being the local electron density, the phase $\phi(\mathbf{r}_A) = \mathbf{k} \cdot \mathbf{r}_A - \omega t + \phi_0(\mathbf{r}_A)$ of the electronic wave function is completely determined by the external light field (Fig. 3A). The wave function oscillates at a frequency of $\sim 10^{14}$ Hz (i.e., optical frequency). Classically, this is a forced oscillation, where electrons have to assume the enforced local phase by the external field, even surviving scatterings and collisions. Here $\phi_0(\mathbf{r}_A)$ represents the phase lag in response to the external field. It is not the initial random phase, which is wiped off by dissipation. The same applies to the electronic wave functions at \mathbf{r}_B in a different (or the same) flake. Because the two wave functions are both coherent with the light wave, they are coherent to each other too. This is a dynamic or ac electron coherence, which is different from the commonly known steady-state or dc electron coherence in transport measurements. The electrons do not move far away from their equilibrium positions. After the wind-chime formation the electron coherence becomes stronger because more electrons get correlated and fewer collisions get involved, owing to the aligned geometry. At the far field, the intensity is $I(\mathbf{r}_C) = E(\mathbf{r}_C) \cdot E(\mathbf{r}_C)$, where $E(\mathbf{r}_C)$ is the electric field of the light wave at \mathbf{r}_C . It becomes $E(\mathbf{r}_C) = \zeta(\chi^{(3)}) \Psi(\mathbf{r}_C)$, with $\Psi(\mathbf{r}_C) = e^{i(\mathbf{k} \cdot \mathbf{r}_C - \omega t + \phi_0)} [\sqrt{\rho_A(\mathbf{r}_A)} e^{-i\mathbf{k}_\perp \cdot \mathbf{r}_A} + \sqrt{\rho_B(\mathbf{r}_B)} e^{-i\mathbf{k}_\perp \cdot \mathbf{r}_B}]$ being the optical phase except for a normalization factor. Here $k_\perp = d\Delta\phi/dr$ is the wavevector generated due to the nonlinear response term of n_2 (Methods). The exact coefficient $\zeta(\chi^{(3)})$ is determined by the optical nonlinearity $\chi^{(3)}$, which is comprehensively determined by the carrier properties in the material.

We found that SSPM can be used to measure easily the $\chi_{\text{onelayer}}^{(3)}$ value for many of the layered quantum materials (we have also observed SSPM in dispersion of MoSe₂ flakes and

obtained the $\chi^{(3)}$ value). In other coherent methods (e.g., THG or FWM), the signal is much weaker than the input beam (25). Instead, in SSPM the diffracted beam is strong, owing to the intrinsic electron coherence. Significantly, SSPM can be used to measure $\chi^{(3)}$ from near IR to UV, compared with THG working only for the region where the 3ω signal can be easily detected. This is essential for materials with a relatively large gap. In this sense we have found a powerful and ubiquitous (in some cases unique) way to measure $\chi^{(3)}$ for many of the 2D gapped quantum materials.

To quantitatively investigate the gap-dependent property, we obtained $\chi^{(3)}$ values for excitations at multiple wavelengths (Fig. 4A). For example $\chi_{\text{onelayer}}^{(3)}$ at 532 nm is 1.6×10^{-9} electrostatic units (e.s.u.) (i.e., 2.23×10^{-17} m²/V²; Methods), which is orders of magnitude larger than that of conventional semiconductors, owing to the high carrier density and mobility in 2D layered materials. In parallel we calculated the absorption as a function of wavelength for few-layer MoS₂, where the dipole transition is proportional to the occupation probabilities of valence and conduction bands. As shown in Fig. 4A, the experimental results and theoretical calculations compare well. A sharp change in $\chi^{(3)}$ and threshold values can be seen at 1.72 eV (722 nm), which is exactly the gap value of MoS₂. This gap-dependent property is a manifestation of the electronic band structure of MoS₂. Thus, SSPM can be used to investigate band structures of layered quantum materials and can be conveniently compared with other experimental methods such as absorption and photoluminescence. The SSPM method applies especially to those materials where large single crystals are not available and absorption is hard to measure.

In Fig. 4A the experimental results compare well with the calculated absorption along x and y , but not that along the z direction (SI Text, section S2). This is strong evidence that, in our experiment, light propagation is perpendicular to the x or y but not the z axis, which confirms the validity of the wind-chime model. Note that this is an even stronger restriction than the wind-chime model.

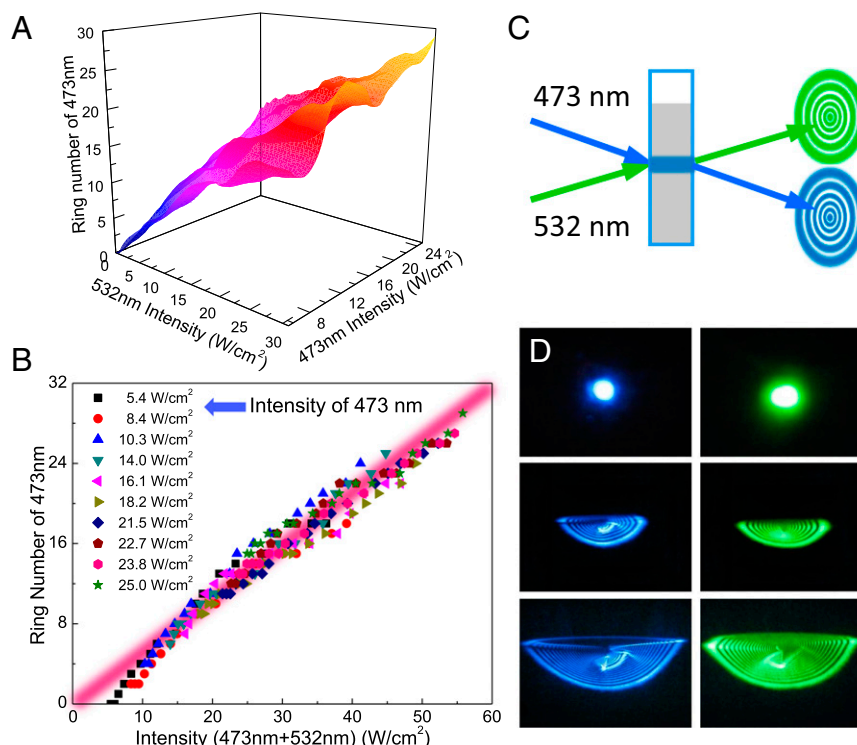


Fig. 5. All-optical switching based on SSPM. (A and B) Dependence of the 473-nm beam ring number on the sum intensity. (C) Schematic of the switch. (D) Generating the 473-nm pattern by increasing the 532-nm beam intensity. Both ring numbers (and diameters) increase simultaneously, with fixed 473-nm intensity below threshold.

($f = 200$ mm) and the center of the cuvette is kept fixed at 150 mm. The beam spot size after focusing is 0.2 mm for the $1/e^2$ intensity radius. In the cw experiment, single transverse-mode 532-nm and 473-nm laser beams were used instead, having a similar beam spot size on the sample. The relation between the fringe thickness and the flake concentration is investigated and discussed in [SI Text, section S4](#). Micrometer-sized powder of MoS₂ crystals (LSKYD, [www.kydmaterials.com/en/index.php](#)) was added into the solvent and then ultrasonicated for 30 min before the measurement. The optimum concentration is 0.14 g/L (i.e., 8.5×10^{-4} mol/L). From our Raman and absorption characterization there are very few monolayer MoS₂ flakes in our sample ([SI Text, section S5](#)). From Fig. 1B, it can be seen that our sample is mainly composed of MoS₂ flakes with a thickness of 30 ~ 55 layers. Being different from the linear optical phenomena of Newton's rings, the SSPM pattern has a finite number of fringes and the thickest fringe has the largest diameter. The critical proof of observing a SSPM pattern is that the number of rings, N_r , increases linearly with the input laser intensity.

Fundamentals of SSPM and the Calculation of the $\chi_{\text{onelayer}}^{(3)}$ in MoS₂. The nonlinear optical response SSPM is characterized by $n = n_0 + n_2 I$, where n_0 and n_2 are the linear and nonlinear refractive indexes, respectively. Assuming a Gaussian beam, the optical phase accumulated after propagation length l is $\Delta\psi(r) = (2\pi n_0/\lambda) \int_{-l/2}^{l/2} n_2 I(r, z) dz$, where z is the propagation direction of the incident laser beam. Thus, we have $\Delta\psi(0) - \Delta\psi(\infty) = (2\pi n_0/\lambda) n_2 \cdot 2l$, with $I(0, z) = 2I$. The number of rings N is proportional to the phase change in the output beam ($\Delta\psi(0) - \Delta\psi(\infty) = 2\pi N$), leading to $n_2 = (\lambda/2n_0 l)(N/l)$, where $n_0 = 1.36$ and N is proportional to l for a nearly constant n_2 . By the slope in Fig. 2 we can straightforwardly obtain n_2 from the experimental data. With $n_2 = (12\pi^2/(n_0^2 c)) \cdot 10^3 \chi^{(3)}$ (in the SI unit) we estimate that $\chi^{(3)} = 1.44 \times 10^{-4}$ e.s.u. for the 532-nm cw laser beam excitation. The number of effective single layers, M , that the laser beam passes through is estimated to be about 300, using the concentration of the sample (*SI Text, section S6*). As $l_{\text{total}} \sim M^2 l_{\text{onelayer}}$,

we estimate that $\chi^{(3)} = M^2 \chi_{\text{onelayer}}^{(3)}$. The estimated $\chi_{\text{onelayer}}^{(3)}$ for 532 nm is 1.6×10^{-9} e.s.u. (i.e., 2.23×10^{-17} m²/V²).

Conveyance of the ac Electron Coherence to the Optical Coherence. The diffraction of the beam off the z axis is due to a finite perpendicular wavevector, which is, for a Gaussian beam $\Delta\psi(r) = \Delta\psi(0) \exp(-2r^2/a^2)$, explicitly written as $k_{\perp} = d\Delta\psi(r)/dr = (-16\pi n_0 n_2 l / a^2) r \cdot \exp(-2r^2/a^2)$. After the wind chime is formed, the optical coherence forming the fringes is conveyed from the electron coherence:

$$\begin{aligned} \Psi(\mathbf{r}_C) &= \left[\sqrt{\rho_A(\mathbf{r}_A)} e^{i\phi_0(\mathbf{r}_A)} e^{i|\mathbf{k}_C \cdot (\mathbf{r}_C - \mathbf{r}_A)} e^{i \left[\nabla_{r_\perp} (\Delta\phi) \right]_{r_A}} \right] \widehat{r_\perp} \cdot (\mathbf{r}_C - \mathbf{r}_A) \\ &\quad + \sqrt{\rho_B(\mathbf{r}_B)} e^{i\phi_0(\mathbf{r}_B)} e^{i|\mathbf{k}_B \cdot (\mathbf{r}_C - \mathbf{r}_B)} e^{i \left[\nabla_{r_\perp} (\Delta\phi) \right]_{r_B}} \right] \widehat{r_\perp} \cdot (\mathbf{r}_C - \mathbf{r}_B) + \dots \\ &= e^{i(\mathbf{k}_C \cdot \mathbf{r}_C - \omega t)} \left[\sqrt{\rho_A(\mathbf{r}_A)} e^{i\phi_0(\mathbf{r}_A)} e^{i \left[\nabla_{r_\perp} (\Delta\phi) \right]_{r_A}} \right] \widehat{r_\perp} \cdot (\mathbf{r}_C - \mathbf{r}_A) \\ &\quad + \sqrt{\rho_B(\mathbf{r}_B)} e^{i\phi_0(\mathbf{r}_B)} e^{i \left[\nabla_{r_\perp} (\Delta\phi) \right]_{r_B}} \right] \widehat{r_\perp} \cdot (\mathbf{r}_C - \mathbf{r}_B) + \dots \\ &= e^{i(\mathbf{k} \cdot \mathbf{r}_C - \omega t + \phi_0)} \left[\sqrt{\rho_A(\mathbf{r}_A)} e^{-i\mathbf{k}_\perp \cdot \mathbf{r}_A} + \sqrt{\rho_B(\mathbf{r}_B)} e^{-i\mathbf{k}_\perp \cdot \mathbf{r}_B} + \dots \right]. \end{aligned}$$

Here we have taken that ϕ_0 is identical for r_A and r_B , and only identical k_\perp can produce interference rings.

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