## **Proposal for a Quantum-Degenerate Electron Source**

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We propose a pulsed electron source capable of 6D brightness orders-of-magnitude greater than that of existing sources. It could deliver average current up to 0.5 pA and achieve an emittance approaching the quantum limit in each spatial dimension. It could be employed to advantage in electron microscopy, inverse photo-emission, precision low-energy scattering experiments, and electron holography. This source could make possible pump-probe experiments with Ångstrom spatial and subpicosecond time resolution. Here, we present the basic concepts of the source, including a generalized expression for the brightness that can be used in the quantum limited case and the analysis of the main issues that must be addressed for successful construction and operation. We have begun an experiment to demonstrate its essential features.

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Progress in the fields of electron microscopy, inverse photo-emission, precision low-energy electron scattering experiments, and electron holography strongly depends on the brightness of available electron sources. The conventional definition of brightness B is

$$B = \frac{N_e}{\epsilon_x \epsilon_y \epsilon_z},\tag{1}$$

where  $N_e$  is the number of particles and  $\epsilon_i$  are the emittances with i = x, y, z [1]. For the case of nonrelativistic particles, it is convenient to use  $\beta_{x,y,z}$ , the particle velocity components in speed of light units, as the conjugates of the spatial coordinates. In this frame, quantum mechanics defines the elementary phase space volume as  $\lambda_c^3$ , where  $\lambda_c$  is the reduced Compton wavelength ( $\approx 3.86 \times 10^{-11}$  cm for the case of electrons). By expressing the emittances in (1) in  $\lambda_c$  units, one obtains a dimensionless brightness  $b_F$  that represents the average number of particles in the elementary phase-space cell:

$$b_F = B\lambda_c^3. \tag{2}$$

In the case of electrons, the Pauli principle states that  $b_F$  must be smaller than 2 or 1 for the unpolarized or polarized cases, respectively.

At present, the brightest sources employ field emission from needles or carbon nanotubes and achieve  $b_F = 5 \times 10^{-6} - 5 \times 10^{-5}$  [2,3]. Field enhancement near the needle permits extraction of a large current from a very small area, and this reduces phase volume, making these sources two or three orders of magnitude brighter than those of the previous generation. However, this increase in brightness is limited by electron-electron (interbeam) scattering at high current densities, which adversely affects both longitudinal and transverse emittance [4], and as a result, only 1 in ~10<sup>5</sup> elementary cells is actually occupied by an electron.

Recently, there have been two proposals [5,6] to increase brightness by employing large emission areas with very cold electrons. This became possible with development of

lasers and laser-cooling techniques (bright atomic beams and optical molasses). Both proposals discuss pulsed sources and near-threshold photo ionization or excitation of high level Rydberg states of Cs, which can result in production of cold electrons. In [6], the source is intended for an accelerator and requires a large number of electrons per bunch. In this case, the brightness is limited, at best, by an effective temperature  $kT_k \sim e^2 n_e^{1/3}$  (where k is the Boltzmann constant,  $T_k$  is the temperature, and e and  $n_e$ are the electron charge and density, respectively).  $T_k$  is related to fluctuations in the potential energy in the random distribution of ionized atoms. The authors expect to achieve a factor of 10 improvement compared to the best existing sources for accelerators, which obtain  $b_F \sim 10^{-11}$ . In [5], to avoid problems with scattering and random positioning of electrons, our group proposed a source that operates in pulsed mode where on average only one electron per pulse is generated. For such a source suitable, for example, for pump and probe experiments and scanning microscope applications, a value of  $b_F \sim 2 \times 10^{-3}$  was predicted.

In this Letter, we extend and improve the concepts presented in [5] and describe an electron source capable of nearly achieving the brightness quantum limit. In the first part, we introduce a generalized expression that allows proper representation of brightness in the quantum regime, while in the second part, we describe the source scheme and the experimental requirements for the source construction.

When  $b_F$  approaches unity, the wave property of the electron becomes more and more important, and brightness, which in the classical case is invariant as a consequence of Liouville's theorem, is no longer conserved [7]. In fact, in wave optics, phase volume is not conserved, and as an example, we can consider the case of a beam of coherent photons occupying a certain volume in phase space (as generated, for example, by a laser). By using a beam splitter, we can separate the beam into 2 parts, and

the total phase space occupied by the coherent photons will now be twice the original while the brightness will be half the original. However, by using a Michelson interferometer, we can recombine the two beams and restore the initial conditions exactly.

The invariant in wave optics is no longer the brightness, but is instead the degeneracy parameter  $\delta$  defined as the number of particles crossing the transverse coherence area in longitudinal coherence time [8]. It follows that for any pure quantum state of polarized electrons, the degeneracy parameter is equal to unity. In all real situations, including that of our proposed source, one does not have a pure quantum state, because one or more of the parameters describing the system fluctuate randomly. In this case, one must employ the density matrix (the quantum analog of the density in phase space for the classical physics of particles). When the parameter fluctuations become sufficiently strong, the off-diagonal elements of the density matrix become negligibly small, and one arrives at the limit where the classical physics of particles is appropriate, and the brightness as expressed by (1) or (2) recovers its meaning.

However, for the source we are proposing, the parameter fluctuations are relatively small, and in order to give an adequate description of such a case, we must express the degeneracy parameter  $\delta$  previously defined in quantum mechanical terms.

As a preliminary, we introduce the operational scheme for our source. A beam of neutral alkali-metal atoms interacts in a small volume V with the photons from a pulsed laser system, and on average, only one atom per pulse is excited into a band of high lying Rydberg nPstates. The radial motion of the resulting valence-electron Rydberg wave packet is nearly classical. Conditions are chosen so that for every pulse at apogee, the valence electron has very nearly the same spatial wave function, which is shell-like with radius R and thickness  $\Delta R \ll R$ . When the electron reaches apogee, an electric field pulse is applied that ionizes the atom and accelerates the electron away from its parent ion. Before the next electron generation cycle, an additional electric field pulse removes the parent ion from the interaction region.

In full generality, in order to define  $\delta$  in quantum mechanical terms, one should start by constructing the density matrix for *N* excited atoms. However, as an adequate approximation, it is sufficient to consider just two identical atoms, where one nucleus is displaced from the other by distance  $\vec{u}$  and has a relative "velocity"  $\vec{\beta}$ . (We ignore the very small uncertainty  $\Delta u_i \Delta \beta_i \sim \hbar/Mc$ , where *M* is the alkali-metal atomic mass,  $\hbar$  the reduced Planck constant, and i = x, y, z). We assume that  $\vec{u}$  and  $\vec{\beta}$  have the normalized probability distributions  $j(\vec{u}), J(\vec{\beta})$  about their respective origins with standard deviations  $\vec{\sigma}, \vec{\eta}$ , respectively. Let the normalized valence-electron wave function at apogee be  $\psi(\vec{r})$  when  $\vec{u} = \vec{\beta} = 0$ . If we neglect Coulomb interaction between electrons, the antisymmetrized two-electron wave function is

$$\Psi(\vec{r}_1, \vec{r}_2) = 2^{-1/2} [\psi(\vec{r}_1)\psi(\vec{r}_2 - \vec{u}) \exp(i\vec{\beta} \cdot \vec{r}_2/\lambda_c) - \psi(\vec{r}_1 - \vec{u})\psi(\vec{r}_2) \exp(i\vec{\beta} \cdot \vec{r}_1/\lambda_c)].$$
(3)

The corresponding density matrix is

$$\rho(\vec{r}_1, \vec{r}_2, \vec{r}_1', \vec{r}_2') = \int j(\vec{u}) d^3 \vec{u} \int J(\vec{\beta}) d^3 \vec{\beta} \Psi(\vec{r}_1, \vec{r}_2) \Psi^*(\vec{r}_1', \vec{r}_2').$$
(4)

The trace of the density matrix is  $Tr(\rho) = 1 - \delta$ , where

$$\delta = \int j(\vec{u}) d^3 \vec{u} \int J(\vec{\beta}) d^3 \vec{\beta} \left| \int \psi \left( \vec{r} + \frac{\vec{u}}{2} \right) \psi^* \left( \vec{r} - \frac{\vec{u}}{2} \right) \right. \\ \left. \times \exp \left( \frac{i}{\lambda_c} \vec{\beta} \cdot \vec{r} \right) d^3 \vec{r} \right|^2.$$
(5)

When  $\vec{\sigma} = \vec{\eta} = \vec{0}, \delta = 1$ . Also, it can be shown that when  $\sigma_i \gg R, \ \eta_i \gg \lambda_c / \Delta R, \ (i = x, y, z),$ 

$$\delta \to \Lambda_c^3 \prod_i 1/(\sigma_i \eta_i).$$
 (6)

Since  $\epsilon_i = \sigma_i \eta_i$ ,  $\delta \to b_F$  in the classical limit. Since the definition of  $b_F$  is to some extent arbitrary, the same is true of  $\delta$ . Several reasonable alternative definitions of  $\delta$  yield results in the classical limit that are very similar but not identical to (6). These will be discussed in detail in a forthcoming publication.

In our source, where we generate on average a single electron per pulse, expression (5) allows us to calculate how small the temperature of the atomic beam and the size of the interaction volume V must be in order to efficiently fill the elementary phase-space cell.

Let us now consider the characteristics of  $\delta$  for our source (see Fig. 1). An atomic beam, Cs in our case, effusing from an oven at temperature ~500 K, can be collimated to reduce the transverse beam temperature to ~5 K, and transverse cooling using the  $6^2S_{1/2}$ - $6P_{3/2}$  Cs resonance line can be employed to further reduce this temperature to ~0.01 K. Also, laser excitation along the



FIG. 1. Schematic diagram of source (not to scale). C: atomic beam collimator; M.S.: magnetic shield;  $L_1$ ,  $L_2$ ,  $L_3$ : Laser beams; V: laser-atom interaction region;  $E_1$ ,  $E_2$ : plane electrodes; A.B.: atomic beam; A: electron extraction aperture. Transverse cooling laser beams not shown.

beam axis can be used to select a narrow band of longitudinal velocities. The Cs atom is excited to a very high lying band of *nP* Rydberg states (for example  $\bar{n} \sim 800$ ,  $\Delta n \sim$ 50), with 3 mutually perpendicular laser beams, L1: cw, 852 nm (6 $S_{1/2}$ -6 $P_{3/2}$ ); L2: cw, 1.47  $\mu$ m (6 $P_{3/2}$ -7 $S_{1/2}$ ); L3: pulsed, 777 nm  $(7S_{1/2}-nP)$ . These laser beams intersect the atomic beam in a small interaction volume  $V \sim (10 \ \mu m)^3$ midway between two plane parallel electrodes E1, E2 with separation  $\sim 1$  cm. The resulting valence-electron Rydberg wave packet forms a spherical shell that expands radially in Kepler-like motion, with half-period  $T = (\pi \hbar^3 \bar{n}^3)/(me^4)$ , where *m* is the electron rest mass. We choose  $\bar{n} \sim 800$ because in this case,  $T \sim 40$  ns, which makes possible a range of pulsed laser bandwidths convenient for chirping, and this, as we will show later, greatly enhances  $\delta$ . However,  $\bar{n} \sim 600$  might also be practical. The oscillator strengths for  $7S-nP_{3/2}$  transitions are much larger than for Cs 7*S*-*nP*<sub>1/2</sub> when  $n \ge 15$  [9]. Taking into account this spin-orbit effect [10] and assuming that the 777 nm photons are linearly polarized along z, the probability density of the Rydberg wave packet is proportional to  $1 + 3\cos^2\theta$ , where  $\theta$  is the angle between the z axis and the electron position vector  $\vec{r}$ .

One wants the shell width  $\Delta R$  at apogee to be reasonably large because this relaxes restrictions on how small the interaction volume V must be. Also, it is desirable that the radial part F of the wave function describing the wave packet at apogee be real (apart from an arbitrary overall phase), so that the radial current density vanishes everywhere. These goals are achieved by suitable chirping of the 777 nm laser pulse. To describe this, it is convenient to measure time in units of T and distance in units of  $R \cong$  $2\bar{n}^2 a_0 \cong 67 \ \mu \text{m}$  in our case ( $a_0$  being the Bohr radius). In these units, the radial wave function at time t = 1 can be expressed as F(r), and the laser field as  $A(t) \exp[i\phi(t)]$ where A(t) and  $\phi(t)$  are the slowly varying amplitude and phase of the laser pulse, respectively. One can show [11] that Kepler's laws and the requirement that F(r) is real uniquely define

$$\phi(t) = -\pi \bar{n} [(3/2)(1-t)^{1/3} + t/2], \tag{7}$$

and yield the following relations between the envelope function A(t) and F(r):

$$A(t) \sim (1-t)^{-1/3} F[(1-t)^{2/3}], \tag{8}$$

$$F(r) \sim r^{1/2} A(1 - r^{3/2}).$$
 (9)

These analytical expressions have been checked by numerical integration of Schroedinger's time-dependent equation for excitation of a Cs atom by a laser pulse given by (7) and (8) assuming a definite F(r), and subsequent evolution to apogee. The calculation reproduces the initial choice of F(r) to very good accuracy regardless of the form of F within wide limits, so long as  $\phi$  is given by (7). The laser pulse shape and chirping parameters can be

properly chosen to generate arbitrary F(r). In Fig. 2, we plot the pulse intensity  $[A(t)]^2$  and the pulse frequency  $\nu = \text{const} - 1.03 \times 10^{10} [3(1-t)^{1/3}(2t)^{-1} + 1/2 - 3/(2t)]$  Hz obtained from (7), assuming

$$F(r) = \operatorname{const} \times \exp[-(r-1)^2/(4\Delta R^2)], \quad (10)$$

with  $\Delta R = 0.12R$  in our case.

Such a choice for F(r) generates a realistic laser pulse, and allows to drastically simplify Eq. (5) from a 12-D to a 1-D integral when Gaussian probability distributions j, Jare assumed for the quantum case where  $\sigma \ll 1$  and  $\eta \Delta R \ll 1$ :

$$\delta = \int_0^1 \frac{\exp(-2\eta^2 x)}{\sqrt{1 + \frac{\sigma^2}{2\Delta R^2} + \frac{\sigma^4}{4\Delta R^4}x(1-x)}} dx.$$
 (11)

In Fig. 3, we plot curves of constant  $\delta$  from (11) for two cases: (a) the chirped laser pulse described by (7)–(10), with (11),  $\bar{n} = 800$  and  $\Delta R = 0.12R$ ; (b) an unchirped Gaussian laser pulse amplitude with time dependence  $\exp(-t^2/4\tau^2)$ . We choose  $\tau = 1$  ns because this gives a wave packet with minimum uncertainty  $\Delta R \Delta p_r$  at apogee as a function of  $\tau$  for  $\bar{n} = 800$ . Here,  $\Delta R \sim 0.02R$ . This unchirped laser pulse yields a radial wave function at apogee with an r-dependent phase factor, and thus a radial probability current density that does not vanish identically. It can be shown that this results in  $\delta$  described by (11) where  $\sigma$  in the integrand is replaced by  $\sigma_{\rm eff} \sim \sqrt{2}\sigma$ . It appears practical to achieve  $\sigma \sim 10 \ \mu m$  and  $\eta c \sim$ 100 cm/s in a real apparatus. Assuming this, Fig. 3 shows that for case (a),  $\delta \sim 0.6$ , while for case (b),  $\delta \sim 0.03$ , revealing the large advantage of chirping.

We next discuss various important considerations for the source operation. At apogee, an electric field pulse with peak value  $\mathcal{E}_0$  and FWHM =  $\tau_0$  is applied for ionizing the atom and accelerating the electron.  $\mathcal{E}_0$  and  $\tau_0$  should satisfy the following criteria: (1)  $\int \mathcal{E}dt$  should be the same for all electrons regardless of  $\theta$  at apogee; (2) the acceleration should be rapid enough that the sudden approximation of time-dependent perturbation theory is valid and Coulomb interaction between the electron and residual Cs<sup>+</sup> ion is minimal (this assures that the shell wave packet is accelerated "intact" and with minimal distortion); and (3) the



FIG. 2 (color online). Laser pulse intensity (solid curve) and change in frequency (dashed curve) for chirped pulse described by Eqs. (7)-(10).



FIG. 3. Curves of constant  $\delta$  in the  $\eta\sigma$  plane, calculated from Eq. (11). Dashed curves: case (a) chirped with  $\Delta R = 0.12R$ . Solid curves: case (b) no chirp,  $\Delta R \sim 0.02R$ .

final electron energy *E* should be sufficiently large (~1–10 eV) that electron-optical aberrations on passing through apertures are not too serious. Numerical simulations reveal that acceptable values are  $\mathcal{E}_0 \sim 100 \text{ V/cm}$ ,  $\tau_0 \sim 0.5 \text{ ns.}$ 

For a Rydberg atom with principal quantum number *n*, the electric field required for spontaneous ionization is  $\mathcal{E}_I = e/(16a_0^2n^4) \sim 8 \times 10^{-4}$  V/cm for n = 800. Such a field is generated by an electronic charge at a distance s = 0.014 cm. Therefore, we must ensure that when the next laser pulse occurs, the residual ion from the present pulse is at a distance  $s' \gg s$  from the interaction region. To achieve this, after the acceleration pulse, another "ion clearing" electric field pulse must be applied to remove the remaining Cs ion from the interaction region. Because the ion is massive, the clearing pulse must have duration ~100 ns if its amplitude is ~1 kV/cm. In this case, a cycle repetition rate ~3 MHz can be achieved. At an average of one electron per pulse, this gives an average source current of ~0.5 pA.

Additional effects due to electric and magnetic stray fields, collisions of Rydberg atoms with ground state Cs atoms and with  $Cs_2$  molecules, and electron photoemission induced by stray laser photons, need to be carefully taken into account in any real device. An analysis of such effects for the case of our source can be found in [11].

In conclusion, we propose a pulsed electron source with nearly quantum limited degeneracy that could be used for pump and probe experiments where nearly full longitudinal and transverse coherence is required. For such applications, the best existing source will have on average  $\sim 5 \times 10^{-5}$  electrons per pulse with the required coherence compared with the 0.5 electrons per pulse of our source. This 4-orders-of-magnitude better performance could open the way to new kind of applications in the coherent imaging field such as electron holography. Successive pulses will not be mutually coherent, but as in any coherence experiment with electrons, interference occurs between different paths over a single electron wave packet. If successive pulse wave packets are similar enough (as in our source), an interference pattern with significant visibility emerges from the train of pulses. The source would also extend applications for electron beams to energy exchange with  $\mu eV$  accuracy in inelastic atomic and molecular scattering, and to new ways of investigating chemical reactions and dynamics on a picosecond time scale using pump-probe techniques. Finally, this source could be the first step toward a novel low-energy scanning electron microscope with current density on target of  $\sim 1 \text{ pA/Å}^2$  (comparable with existing scanning microscopes), but with Ångstrom resolution at few hundreds eV (at the present time, a comparable resolution can be obtained only at many hundreds keV). Such resolution could be achieved for two main reasons: chromatic aberration is minimized by the extremely small phase-space volume, and the well-defined time structure of the electron beam allows for spherical aberration correction in axial symmetry using pulsed electromagnetic optics.

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- C. Lejeune and J. Aubert, *Applied Charged Particle Optic, Advances in Electronics and Electron Physics*, edited by A. Septier, (1980), Supp. 13 A.
- [2] J. C. Spence and M. R. Howells, Ultramicroscopy **93**, 213 (2002).
- [3] N. deJonge, M. Allioux, J. T. Oostveen, K. B. K. Teo, and W. I. Milne, Phys. Rev. Lett. 94, 186807 (2005).
- [4] See for example P. Kruit and G. H. Jansen, *Handbook of Charged Particle Optics* (CRC Press, Cleveland, Ohio, 1997), pp. 275–319.
- [5] M. Zolotorev et al., in Proceedings of the 2005 Particle Accelerator Conference, Knoxville, TN, 2005 (IEEE, New York, 2005), p. 770.
- [6] B.J. Claessens et al., Phys. Rev. Lett. 95, 164801 (2005).
- [7] See for example P. Kruit, M. Bezuijen, and J.E. Barth, J. Appl. Phys. **99**, 024315 (2006).
- [8] See for example J.C.H. Spence, W. Qian, and M.P. Silverman, J. Vac. Sci. Technol. A 12, 542 (1994).
- [9] D. Norcross, Phys. Rev. A 7, 606 (1973).
- [10] H. Kaminski, J. Kessler, and K. J. Kollath, Phys. Rev. Lett. 45, 1161 (1980).
- [11] M. Zolotorev, E. D. Commins, and F. Sannibale, Lawrence Berkeley National Lab. Report No. LBNL-61587, 2006.