Absorption-Limited Generation of Coherent Ultrashort Soft-X-Ray Pulses

M. Schnürer,[1,*] Z. Cheng,[1] M. Hentschel,[1] G. Tempea,[1] P. Kálmán,[2] T. Brabec,[1] and F. Krausz[1]
[1]*Institut Angewandte Elektronik u. Quantenelektronik, Technische Universität Wien, Gusshausstrasse 27-29, A-1040 Wien, Austria*
[2]*Department of Experimental Physics, Budapest University of Technology, Budafoki út 8, F.I.I.10, H1521 Budapest, Hungary*
(Received 30 December 1998)

Coherent growth of high-order harmonic radiation has been extended to propagation lengths comparable to the XUV absorption length at wavelengths as short as 10 nm range for the first time. Absorption-limited production of high harmonics of sub-10-fs near-infrared laser pulses in gas jets gives rise to a harmonic conversion efficiency of $(3-4) \times 10^{-8}$ in the range of 10–13 nm in neon and some 2 orders of magnitude higher at about 30 nm in argon. This kHz-repetition-rate ultrafast soft-x-ray source emits approximately 10^{10} photons per sec within a 0.9-nm bandwidth of a typical Mo:Si mirror at 13.4 nm in a near-diffraction-limited beam and opens up the way to soft-x-ray nonlinear optics and the measurement of sub-fs soft-x-ray pulses.

PACS numbers: 42.65.Ky, 41.50.+h, 42.50.Hz, 42.65.Re

Motivated by a wide range of applications, coherent femtosecond sources have recently been extended to soft x-ray wavelengths of $\leq$10 nm by generating high-order harmonics of intense ultrashort laser pulses in noble gas targets [1–4]. Because the emission of these high-energy photons is inherently coupled with ionization [5,6], the emerging free electrons give rise to an increased phase velocity of the fundamental wave as compared to that of the harmonic wave. This phase-velocity mismatch severely limits the coherent growth of the short-wavelength harmonic radiation, resulting in a conversion efficiency rapidly decreasing with decreasing wavelength.

Recently, precise pressure control in a capillary waveguide along with 20-fs driver pulses allowed compensation of the negative contribution to the refractive index (arising from the free electrons and the waveguide) by the positive contribution of the neutral atoms, resulting in enhanced harmonic conversion efficiencies of greater than 10^{-6} at about 30 nm [7]. Efficiencies of 10^{-7}–10^{-5} in the range of 25–50 nm have also been measured in other recent experiments [8]. By contrast, a fraction of laser light as low as 10^{-11}–10^{-10} could only be converted into individual soft-x-ray harmonics about 10 nm so far [1,9], owing to a lower atomic polarizability and shorter coherence length due to increased ionization at these short wavelengths.

In this Letter we report the generation of *few-cycle-driven* harmonic radiation in the wavelength range of 10–13 nm with efficiencies exceeding by 2 to 3 orders of magnitude, those reported so far. The dramatically shortened rise time of the laser pulse ($\approx$2 optical cycles) as compared to previous experiments allows atomic dipole radiation to be produced at these wavelengths at unprecedentedly low ($<$1%) ionization levels, extending coherent growth of soft-x-ray harmonics for the first time to the limit set by *linear absorption*. We demonstrate absorption-limited harmonic generation in argon at wavelengths about 30 nm and in neon down to $\approx$10 nm. Within the 0.9-nm-wide high-reflectivity band of a (typical) Mo:Si multilayer mirror centered at 13.4 nm, an average photon flux of $\approx 10^{10}$ photons/s is produced in a near-diffraction-limited beam. The source emits bursts of an estimated duration of less than 3 fs at a kHz repetition rate and represents, to the best of our knowledge, the highest-peak-power and highest-average-power coherent laboratory soft-x-ray source demonstrated to date. The achieved peak brightness along with the kHz repetition rate of the source holds promise for inducing the *detectable nonlinear response* of atoms in the soft-x-ray regime and opening the way to *attosecond science*.

The XUV/soft-x-ray harmonic radiation is generated by focusing linearly polarized 7-fs laser pulses carried at $\lambda_0 \approx 790$ nm into an argon and neon gas target. The pulses are delivered by a Ti:sapphire-based laser system up to energies of 0.7 mJ at a 1-kHz repetition rate in a diffraction-limited beam [10]. In the experiments described below the laser has been focused to a $1/e^2$ diameter of $2w_0 \approx 60$ μm or, alternatively, 120 μm, resulting in effective temporal peak intensities (averaged over $A_{\text{eff}} = \pi w_0^2$) up to $I_p = 5 \times 10^{14}$ W/cm^2 and 2×10^{15} W/cm^2, respectively. The atoms are exposed to the laser in a quasistatic gas cell formed by a nickel tube with its axis aligned perpendicularly to the laser propagation direction [4]. The effective interaction length (using tubes with different diameters) has been varied between 0.2 and 8 mm in the experiments [11].

The spectral characterization of the harmonic radiation is performed by a 1-m grazing incidence spectrograph equipped with a 300 grooves/mm platinum-coated grating (248/310G, McPherson). The transmitted signal is detected by an uncoated channeltron (4715G, Galileo) and a lock-in amplifier. Absolute photon yields are measured with an unbiased silicon XUV photodiode (X-UV50C, UDT Sensors) and an electrometer. The photodiode sensitivity follows the simple linear law $N_e = E_{\text{ph}}/3.63$ eV , where N_e is the number of photoelectrons created by a single XUV photon of energy E_{ph}, over the entire XUV

 0031-9007/99/83(4)/722(4)$15.00

and soft-x-ray regime [12]. To block the laser light completely, three aluminum filters are inserted before the photodiode. The filter, originally designed for greater photon energies, consists of a 200-nm-thick aluminum foil protected with 30-nm Al_2O_3 layers on both sides for long-term stability. The calculated transmittivity of the filters has been verified individually using our harmonic source. Experimental data are compared with results obtained from numerical simulations. In our model, the atomic dipole acceleration is calculated using the quantum theory of Lewenstein *et al.* [13] combined with the Ammosov-Delone-Krainov (ADK) ionization rate [14]. Propagation of the laser and XUV pulse in the ionizing gas is computed by numerically solving Maxwell's equations in one spatial dimension.

XUV harmonic emission from argon has been investigated in the parameter range described above. The maximum XUV output obtained by proper setting of the pressure appears to vary within merely a factor of 2, and also the harmonic spectral distribution shows only weak dependence on interaction length and focusing geometry. A typical XUV harmonic spectrum for $L_{\text{eff}} = 3$ mm, $2w_0 = 60$ μm, and $I_p = 2 \times 10^{15}$ W/cm^2 is depicted by the thick solid line in Fig. 1. Harmonic emission reaches its maximum at $p_{\text{opt}} \approx 0.3$ bar. Comparison of the observed emission spectrum with previously reported data from low-density targets [15] and relating the absorption length to the interaction length at p_{opt} ($L_{\text{eff}} > 10L_{\text{abs}}$) reveal that the harmonic yield at wavelengths $\geq$29 nm is limited by absorption in the atomic gas, which rapidly increases for increasing wavelengths beyond 30 nm. The role of absorption has also been confirmed by numerical simulations, which give proper account for the measured pressure dependence of the harmonic yield from a thin ($L_{\text{eff}} \approx 0.4$ mm) target. Owing to the low level of ionization, approximately 3×10^{18} atoms/cm^2 ($L_{\text{eff}} \times$ gas density), yield a harmonic pulse energy and average power in excess of 1 nJ and 1 μW, respectively, at the 27th harmonic. Simulations indicate that the positive contribution of the neutral atoms to the refractive index [7] is not fully compensated by the negative contribution from the free electrons at the intensities where these harmonics first appear on the leading edge of the sub-10-fs driver pulse. This is presumably why maximum emission is achieved at comparatively high intensities (stronger ionization). As a consequence, phase matching can also be achieved with somewhat longer 20–40 fs pulses at these XUV wavelengths [7].

In neon, gentle focusing ($2w_0 = 120$ μm and $I_p = 5 \times 10^{14}$ W/cm^2) gives the best results. Figure 2 depicts harmonic spectra recorded under these conditions for absolute comparison. The lower and middle traces show the emission spectra for $L_{\text{eff}} = 0.2$ mm at pressures of 0.4 and 0.9 bar (maximum yield), respectively. Maximum soft-x-ray output from neon is achieved for effective interaction lengths of 2–3 mm at a pressure of $\approx$0.4 bar. The upper trace depicts the recorded spectrum for $L_{\text{eff}} = 3$ mm and $p = 0.4$ bar. The harmonic signal is enhanced by a factor of 3–4 as compared to the thin target in the range of 10–13 nm. To gain insight into this behavior, we scrutinize contributions to dephasing of the harmonic wave. The ionization level for $I_p = 5 \times 10^{14}$ W/cm^2 reaches $\approx$0.5% during the production of harmonics in the range of 10–15 nm. The contribution of the neutral atoms to the refractive index corresponds to an effective ionization level of $\approx -1\%$. Hence the net pressure-dependent contribution to the refractive index is *positive*, and at sufficiently low pressures can be compensated for by the geometric phase advance, giving rise to an increased coherence

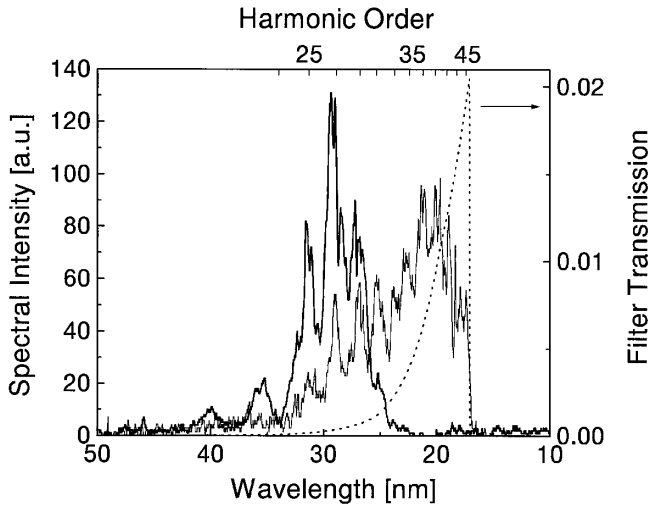

FIG. 1. Spectrum of 7-fs pulse-driven harmonic emission from a 3-mm-long argon gas cell at a pressure of 0.3 bar (thick line) and as transmitted through a couple of aluminum filters (thin line). Dashed line: transmittivity of the two aluminum filters. The sharp cutoff in the spectrum transmitted through the filters indicates that the harmonic spectrum of argon extends below 17 nm.

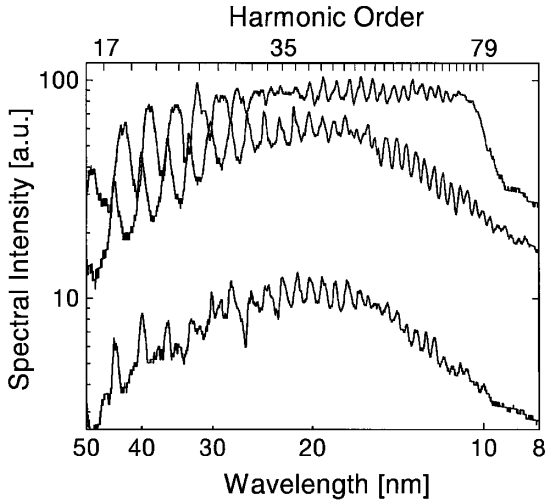

FIG. 2. Spectrum of 7-fs pulse-driven harmonic emission from neon for $L_{\text{eff}} = 0.2$ mm, $p = 0.4$ bar (lower trace); $L_{\text{eff}} = 0.2$ mm, $p = 0.9$ bar (middle trace); $L_{\text{eff}} = 3$ mm, $p = 0.4$ bar (upper trace) for $I_p = 5 \times 10^{14}$ W/cm^2.

length. The low pressure calls for an extended interaction length for maximum yield to be achieved, providing a qualitative explanation for the observed behavior. These considerations have also been corroborated by a comparison of the pressure dependence of the harmonic yield from the two target geometries with numerical simulations.

Quantitatively, the pressure-dependent dephasing length (phase mismatch $= \pi$) for an effective net ionization level of -0.5% is calculated as L_{press} (0.4 bar) ≈ 0.35 mm and the geometric dephasing length [16] is $L_{\text{geom}} \approx 0.6$ mm for the 71st harmonic, whereas the dipole phase [16] evolves notably on a scale of several millimeters and hence can be neglected. A mutual compensation for the two significant dephasing mechanisms could apparently be achieved at a pressure of ≈ 0.25 bar. The need for somewhat higher pressure is presumably due to an ionization level somewhat higher than 0.5% near the optical axis (because I_p is a spatially averaged temporal peak intensity) and to the correspondingly increased value of L_{press}. When the two dephasing effects compensate each other (over a fraction of the laser beam cross sectional area) the coherence length becomes infinite, and absorption comes into play as an effect limiting coherence growth of harmonic radiation. In fact, maximum soft-x-ray harmonic yield (upper trace in Fig. 2) is obtained under conditions resulting in an absorption length 5–10 times shorter as compared to the length of the atomic gas medium. As a result, coherence growth of few-cycle-driven harmonic radiation to the absorption limit gives rise to coherent soft-x-ray emission from some 4×10^{18} atoms/cm^2 in the 10–13 nm range, yielding a conversion efficiency of $(3\text{–}4) \times 10^{-8}$.

To gain further insight into the limiting effects, the power of the 61st harmonic emitted from a thin target ($L_{\text{eff}} = 0.2$ mm) has been measured as a function of the pressure for $2w_0 = 120$ μm and $I_p = 5 \times 10^{14}$ W/cm^2). Geometric and dipole-phase-induced dephasing is small within the interaction region. Defocusing is also found to be weak because the gradient-index lens formed by the free electrons has a focal length more than 3 times longer than the confocal parameter. Hence, the pressure dependence of the signal can be mapped into a dependence on the propagation length at a fixed pressure (0.4 bar) and compared with the computed growth of the harmonic signal in the interaction region (Fig. 3). The measured signal growth rate (squares) is in excellent agreement with the numerical result (solid line) over the full dynamic range of the measurement. With the absorption absent the simulations yield a signal that continues to grow (dashed line) at propagation distances where the measured signal saturates, confirming the limiting role of absorption under our experimental conditions.

Figure 4 shows the efficiency of energy conversion into individual harmonics (within a bandwidth equal to twice the laser frequency) in argon (circles) and neon (squares), in reasonable agreement with the theoretical prediction. The efficiency values have been obtained from measured

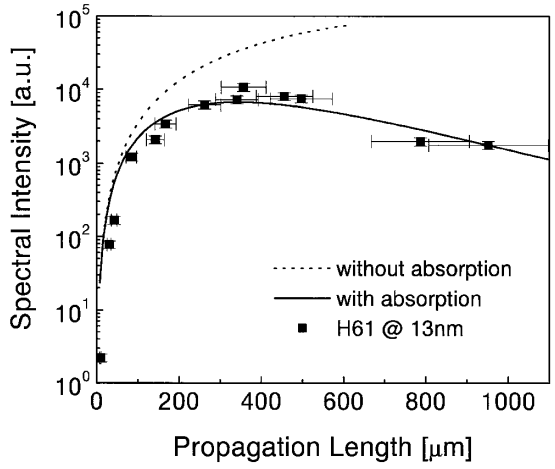

FIG. 3. Measured (squares) and computed (solid line) evolution of the 61. harmonic upon propagation in the neon target for parameters described in the text. The error bars result from uncertainties in estimating L_{eff} and assessing the pressure in the interaction region.

photocurrents of 70 pA in the case of argon and 20 pA in the case of neon. Data below the Al L edge have been obtained from spectral measurements (Fig. 2) by assuming a flat grating-detector response. Within the 0.9-nm high reflectivity band of a typical Mo:Si multilayer mirror centered at 13.4 nm, the demonstrated sub-10-fs-pulse-driven neon harmonic source emits approximately 10^7 photons/pulse within a time interval estimated as $\tau_x \leq 3$ fs from the theoretical model taking into account spatial effects [17]. The enhanced photon yield, together with its unprecedented temporal confinement, results in peak powers of the order of 0.1 MW. These intense soft-x-ray pulses are delivered in a near-diffraction-limited beam (divergence <1 mrad) and hence should be focusable to peak intensities in excess of $I_x = 10^{13}$ W/cm^2.

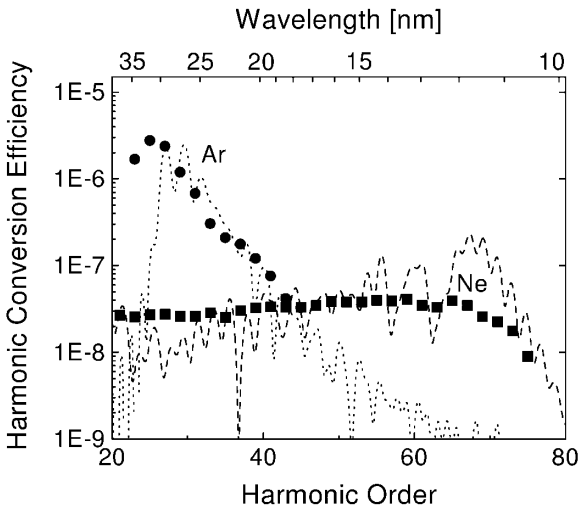

FIG. 4. Harmonic production efficiency in argon, $L_{\text{eff}} = 3$ mm, $p = 0.3$ bar (circles), and in neon, $L_{\text{eff}} = 3$ mm, $p = 0.4$ bar (squares), at $I_p = 2 \times 10^{15}$ W/cm^2 and $I_p = 5 \times 10^{14}$ W/cm^2, respectively, using 7-fs Ti:sapphire laser pulses and calculated spectra, dotted and dashed lines, respectively. The accuracy of the efficiency data is estimated as $\pm 30\%$.

In what follows, we show that these kHz-rate pulses are sufficiently intense for inducing measurable two-photon transitions in the x-ray regime. Following Kálmán [18], the cross section for a two-photon-induced transition from an inner shell of an atom to a free state can be approximated as

$$\sigma^{(2)} \approx \sigma^{(1)}\left(1 - \frac{|E_B|}{2\hbar\omega_x}\right)\frac{3e^2E_x^2a_B}{10(\hbar\omega_x)^3} = 0.34\sigma^{(1)}\left(1 - \frac{|E_B|}{2\hbar\omega_x}\right)10^{-12}\frac{I_x}{(\hbar\omega_x)^3}, \quad (1)$$

where $\sigma^{(1)}$ is the one-photon cross section at the absorption edge, e is the electron charge, a_B is the Bohr radius, E_B is the electron binding energy, and E_x, I_x, and $\hbar\omega_x > E_B/2$ are the electric field strength, intensity, and photon energy, respectively, of the incident x-ray radiation. The second expression yields $\sigma^{(2)}$ in units of cm^2 if I_x is given in $\mathrm{W/cm}^2$ and $\hbar\omega_x$ in eV. Photons of $\hbar\omega_x = 95$ eV (13 nm) are sufficiently energetic to create a K shell vacancy by means of two-photon ionization in boron, for which $E_B = 188$ eV, $\sigma^{(1)} = 9.1 \times 10^{-19}$ cm^2 [19]. For these values, Eq. (1) yields $\sigma^{(2)} \approx 4 \times 10^{-26}$ cm^2 at $I_x = 10^{13}$ $\mathrm{W/cm}^2$. The vacancies are created at a rate of $dN_v/dt \approx N_i f_l \tau_x \sigma^{(2)} I_x/\hbar\omega_x$, where N_i is the number of interacting atoms and f_l is the laser repetition rate. In a volume confined by the area of the focused x-ray beam and the absorption length at ω_x, we have $N_i \approx 10^{11}$, yielding $dN_v/dt \approx 10^4/\mathrm{s}$. Approximately 0.1% of these vacancies are filled upon emitting an x-ray fluorescence photon of energy of $\approx E_B$ and $\approx$99.9% by ejecting an Auger electron [19], both of which should be detectable by state-of-the-art single-photon-counting energy dispersive spectrometry [20] and time-of-flight electron spectrometry [21], respectively. These nonlinear x-ray spectroscopies will allow the temporal characterization of ultrashort soft-x-ray pulses down to the subfemtosecond regime, which cannot be accessed by previously demonstrated techniques [21].

In conclusion, we have reported absorption-limited high harmonic generation down to the 10-nm region, resulting in the highest average and peak powers available from a coherent laboratory source in this wavelength range to date. With a flat-top-like laser beam profile the demonstrated source should be capable of generating megawatt-peak-power sub-fs soft-x-ray pulses, which will be beneficial to traditional (linear) x-ray optics and pave the way towards new (nonlinear and ultrafast) x-ray science.

Fruitful discussions with M. Murnane, H. Kapteyn, M. Drescher, G. Reider, and Ch. Spielmann, as well as the manufacturing of the aluminum filters by K. Ferencz, are gratefully acknowledged. This work has been sponsored by the Austrian Science Fund, Grants No. Y44-PHY and No. P12631-PHY, and by the Ministry of Science and Transportation.

Note added.—Since the submission of this paper, several papers related to our work have been published. Tamaki *et al.* [22] reported coherent emission from some 10^{18} Ne-atoms/cm^2 at 16–32 nm with highly enhanced efficiency. This result can be reconciled with our measurements only by assuming that individual Ne atoms driven by 100-fs pulses radiate much more efficiently as compared to few-cycle-driven harmonics. We were unable to confirm this tendency in our numerical and experimental investigations. Constant *et al.* [23] reported highly efficient absorption-limited harmonic generation in the 40–60 nm range. Our results appear to be in good agreement with the analytic predictions presented in this paper.

*Email address: schnuer@iaee.tuwien.ac.at

[1] A. L'Huillier and P. Balcou, Phys. Rev. Lett. **70**, 774 (1993).
[2] J. J. Macklin, J. D. Kmetec, and C. L. Gordon III, Phys. Rev. Lett. **70**, 766 (1993).
[3] Z. Chang *et al.*, Phys. Rev. Lett. **79**, 2967 (1997).
[4] Ch. Spielmann *et al.*, Science **278**, 661 (1997).
[5] J. L. Krause, K. J. Schafer, and K. C. Kulander, Phys. Rev. Lett. **68**, 3535 (1992).
[6] P. B. Corkum, Phys. Rev. Lett. **71**, 1994 (1993).
[7] A. Rundquist *et al.*, Science **280**, 1412 (1998).
[8] T. Ditmire *et al.*, Phys. Rev. A **51**, R902 (1995); D. Garzella *et al.*, in *Ultrafast Phenomena XI*, edited by T. Elsaesser *et al.*, (Springer, Heidelberg, 1998), p. 387; G. Sommerer, H. Rottke, and W. Sandner, Laser Phys. **9**, 1 (1998).
[9] C.-G. Wahlström *et al.*, Phys. Rev. A **48**, 4709 (1993); K. Kondo *et al.*, Phys. Rev. A **47**, R2480 (1993); G. Sommerer *et al.*, Opt. Commun. **146**, 347 (1998).
[10] S. Sartania *et al.*, Opt. Lett. **22**, 1562 (1997).
[11] L_{eff} is made up of the length of the squeezed tube target and the interaction length outside the tube. The density distribution of the atomic gas streaming out of the gas cell has been calculated by drawing on the relevant discussion in *Atomic and Molecular Beam Methods*, edited by G. Scoles (Oxford University Press, New York, 1988), Vol. 1, Chap. 2, pp. 14–25.
[12] Product catalogue of UDT sensors.
[13] M. Lewenstein *et al.*, Phys. Rev. A **49**, 2117 (1994).
[14] M. V. Ammosov, N. B. Delone, and V. P. Krainov, Sov. Phys. JETP **64**, 1191 (1986).
[15] J. Zhou *et al.*, Phys. Rev. Lett. **76**, 752 (1996).
[16] P. Salieres *et al.*, Phys. Rev. Lett. **74**, 3776 (1995).
[17] In fact, our one-dimensional model predicts the emergence of a single x-ray burst of sub-fs duration, which is broadened by the spatial intensity variation of the laser beam.
[18] P. Kálmán, Phys. Rev. A **39**, 3200 (1989).
[19] R. B. Firestone and S. S. Shirley, *Table of Isotopes* (Wiley, New York, 1996), 8th ed.
[20] M. Schnürer *et al.*, Phys. Rev. Lett. **80**, 3236 (1998).
[21] T. E. Glover *et al.*, Phys. Rev. Lett. **76**, 2468 (1996); A. Bouhal *et al.*, J. Opt. Soc. Am. B **14**, 950 (1997); Y. Kobayashi *et al.*, Opt. Lett. **23**, 64 (1998).
[22] Y. Tamaki *et al.*, Phys. Rev. Lett. **82**, 1422 (1999).
[23] E. Constant *et al.*, Phys. Rev. Lett. **82**, 1668 (1999).