Observation of Quantum Oscillations in Single Crystal Bi Nanowires

Leonid KONOPKO[1,2]
[1]Institute of Electronic Engineering and Industrial Technologies, Academy of Sciences of Moldova
[2]International Laboratory of High Magnetic Fields and Low Temperatures, Wroclaw 53-421, Poland
leonid.konopko@iieti.asm.md

***Abstract* — We present here experimental measurements of Aharonov-Bohm (AB) oscillations on single Bi nanowires with diameter *d* < 80 nm. The single nanowire samples were prepared by Ulitovsky technique and represented cylindrical single crystals with (10$\underline{1}$1) orientation along the wire axis. Due to semimetal-to-semiconductor transformation and big density of surface states Bi nanowire (d ~ 50 nm) effectively turn into conducting tube. The oscillations of longitudinal magnetoresistance (MR) of Bi nanowires with two periods ΔB_1 and ΔB_2 proportional to Φ_0 and $\Phi_0/2$ were observed, where Φ_0=h/e is the flux quantum. From B ~ 8 T down to B = 0 the extremums of $\Phi_0/2$ oscillations are shifted up to 3π at B = 0 which is the manifestation of Berry phase shift due to electron moving in nonuniform magnetic field. Derivative of MR was measured at various inclined angles. The observed angle variation of the periods are not in the agreement with theoretical dependence ΔB(α)= ΔB(0)/cosα of the size effect oscillations of the "flux quantization" type. Moreover the equidistant oscillations of MR exist in transverse magnetic fields under certain rotation angles. An interpretation of the MR oscillations is presented.**

***Index Terms* — Aharonov-Bohm oscillations, Berry phase, bismuth, nanowire, semimetal-to-semiconductor transformations.**

I. INTRODUCTION

Mesoscopic systems have been investigated for many decades. At present they have become the focus of intense experimental and theoretical investigation due to of their scientific and technologic interest. The most exciting opportunity is that of an ideal quantum wire of a diameter d that is less than the Fermi wavelength and with the Fermi level chosen such that the nanowire transport is controlled by a single conduction channel. The properties of this one-dimensional system or quantum wire have been investigated theoretically, the case of Bi nanowires was studied by Hicks and Dresselhaus [1].

Bismuth is a particularly favorable material to study the electronic properties of quantum wires due to its small electron effective mass and high carrier mobility.[2] De Haas–van Alphen and Shubnikov–de Haas effects [3] provides an unambiguous measure of the charge density and the anisotropy of the Fermi surface (FS) of bulk crystalline Bi,[4] which consists of three electron pockets at the *L* point and a *T*-point hole pocket. SdH oscillations also have been studied in single crystal films of Bi [5]. The effective band overlap energy, E_0 , and the Fermi energy, E_F , are 37 and 26 meV, respectively, levels which result in small electron and hole densities (n_i=p_i=3x10^{17} cm^{-3} at a temperature of 4 K). Quantum confinement effects, which decrease E_0, become relevant for quantum wires with diameter $d \approx 2\hbar/\sqrt{2m^*E_0}$, where m^* is the corresponding electron in-plane effective mass transverse to the wire axis. For wires oriented along C_3 (the trigonal direction), m^*=0.0023 and the relevant diameter is 42 nm. Detailed calculations [2], which assume that the components of effective mass tensors do not change upon confinement, show that a semimetal-to-semiconductor (SMSC) transition occurs for d_c~55 nm for wires oriented along the trigonal direction. Various experimental results support this theory. [6]

Quantum interference effects are present in superconducting devices and in very small pure metallic rings and cylinders. In particular, in the presence of magnetic flux Aharonov-Bohm (AB) oscillations [7] may occur in doubly connected systems. [8-10] For a normal metal the period of these oscillations is proportional to Φ_0=h/e (the flux quantum). Such effects should vanish once the elastic mean free path of the electrons is smaller than the system's size. For the disordered cylindrical samples with short mean free path (compare with the circumference of the cylinder) the new type of AB oscillations with period $\Delta B=\Phi_0/2S$ was predicted by Al'tshuler, Aronov and Spivak (AAS) [11]. This effect arises from the interference of pairs of coherent electron waves circumscribing the cylinder. These oscillations were observed by Sharvin and Sharvin [12] on the Mg cylinder 1 μm in diameter and 1 cm long.

AB and AAS oscillations have been observed in various conducting rings,[13-16] tubes,[17] solid cylinders that assimilate tubes such as multiwall carbon nanotubes (MWNTs), [18] Bi nanowire arrays 270 nm in diameter, [19] and single bismuth nanowires in the diameter range of 200–800 nm. [20, 21] In the latter case, theoretical studies have focused on a whispering gallery model of low-effective-mass electrons that define a highly conducting tube in the boundary of the solid cylinder. [22] New type of AB oscillations have been observed in single Bi nanowires with diameter *d* < 100 nm [23, 24].

The present paper is devoted to an experimental investigations of MR oscillations, equidistant in the direct magnetic field, of the Bi cylindrical single crystals with diameter d < 80 nm.

II. EXPERIMENTAL

Individual Bi nanowires were fabricated using the Ulitovsky technique (see schematic diagram at inset on Fig. 1.), by which a high-frequency induction coil melts a 99.999%-pure Bi boule within a borosilicate glass (Pyrex) capsule, simultaneously softening the glass. Glass capillaries containing Bi filament [25] were produced by drawing material from the glass. Encapsulation of the Bi filament in glass protects it from oxidation and mechanical stress.

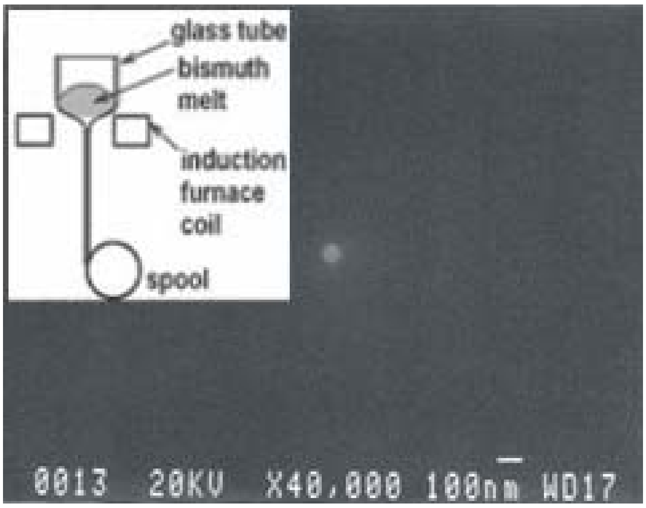

Fig. 1. Scanning electron microscope cross sections of the 55 nm Bi wire in its glass coating. The inset illustrates the Ulitovsky method for synthesizing long, small-diameter wires in a glass fiber.

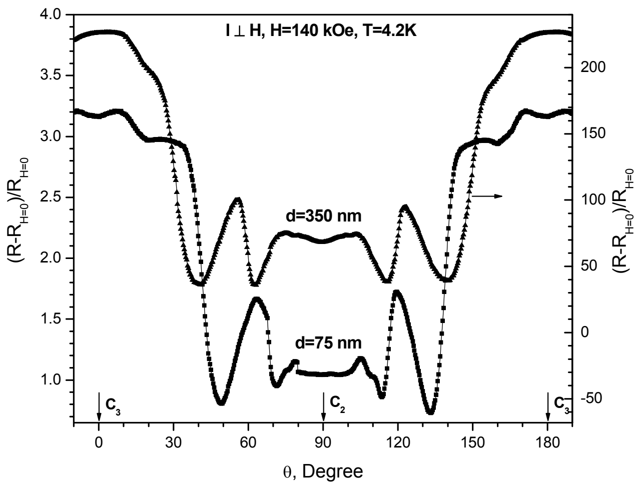

Fig. 2. The angular diagram of transverse magnetoresistance ΔR/R of Bi wires with diameter d=350 nm and d=75 nm at 4.2 K, H=140 kOe.

Electrical connections to the nanowires were performed using $In_{0.5}Ga_{0.5}$ eutectic. This type of solder consistently makes good contacts, as compared to other low-melting-point solders, but it has the disadvantage that it diffuses at room temperature into the Bi nanowire rather quickly. Consequently, the nanowire has to be used in the low-temperature experiment immediately after the solder is applied. The samples used in this work are, to date, the smallest diameter single Bi wires for which the electronic transport at low temperatures has been reported. The wires are characterized by an electronic diameter $d = \sqrt{\rho_{Bi} l / R_0}$, where ρ_{Bi}=1.14x10^{-4} Ohm*cm is the 300 K bulk resistivity, l is the wire length, and R_0 is the wire resistance at T=300 K. We used samples with 45 nm<d<800 nm. A scanning electron microscope (SEM) images of the 55 nm nanowires are shown in Fig. 1. SdH oscillations are observed in the thick (d>150 nm) Bi nanowires showing that the bismuth material in the nanowires has the required high purity and high mobility. However, in the thin samples (d<80 nm) the conditions for observing SdH are not optimal because of the large AB oscillations. As shown in Fig. 2 the resistance changes when the nanowire is rotated around its axis in transverse magnetic field. The rotational diagram of thin nanowires (d<80 nm) are similar to the one shown by large diameter nanowires, with minima and maxima separated by 90°. It has been observed that large diameter (d>150 nm) individual nanowires are single crystals whose crystalline structure, as determined by Laue x-ray diffraction and SdH methods, is oriented with the (10$\underline{1}$1) along the wire axis. In this orientation the wire axis makes an angle of 19.5° with the bisector axis C_3 in the bisector-trigonal plane also, the trigonal axis C_3 is inclined to the wire axis at an angle of ~70°, and one of the binary axes C_2 is perpendicular to it. Therefore, it is reasonable to assume that all nanowires are single crystals. Fig. 3 shows the orientation of our nanowires with respect to the Fermi surface. The electron pockets *LB* and *LC* are located symmetrically with respect to the wire direction.

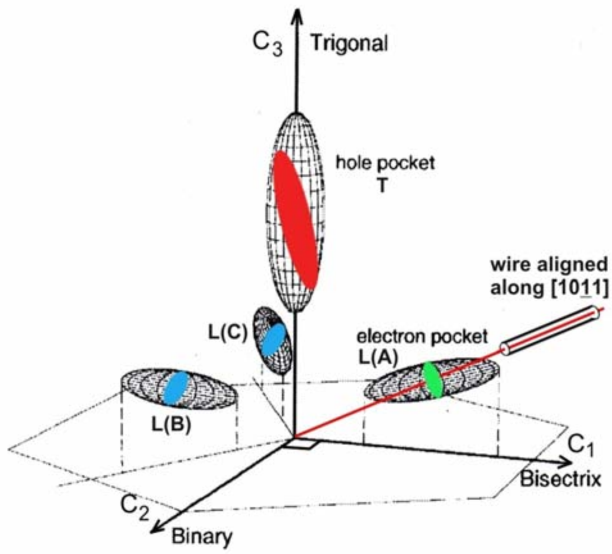

Fig. 3. Schematic diagram of (10$\underline{1}$1) nanowires orientation with respect to the Bi Fermi surfaces of electrons and hole.

III. RESULT AND DISCUSSION

Fig. 4 shows the magnetic field dependence of the longitudinal MR for Bi nanowires d= 55 nm and 73 nm.

R(B) decreases for increasing magnetic field it is typical

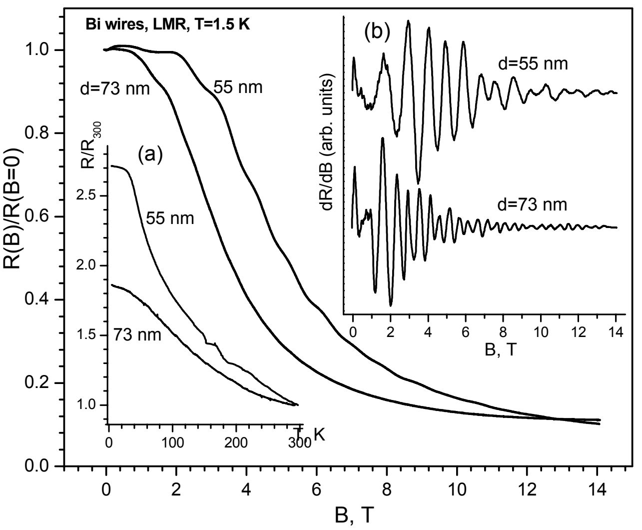

Fig. 4. Magnetic field dependence of the longitudinal MR for a 55 nm and 73 nm Bi nanowires, T=1.5 K. Insert (a): Temperature dependences of the resistance for a 55 nm and 73 nm Bi nanowires. Insert (b): Magnetic field dependence of the derivative of longitudinal MR for 55 nm and 73 nm Bi nanowires, T=1.5 K (the monotonic part is subtracted).

for Bi nanowires of large diameter. [25] This effect has been observed in many studies and by many groups in almost all samples of Bi nanowires even those of small diameter. [2, 26] This phenomenon is a Chambers effect, [27] which occurs when the magnetic field focuses electrons toward the core of the wire (away from the surface), thereby avoiding surface collisions. We have observed the non monotonic changes of longitudinal MR that are equidistant in the magnetic field. The insert (a) in Fig. 10 presents the temperature dependence of resistance R_T/R_{300} for Bi nanowires d= 55 nm and 73 nm. The R(T)

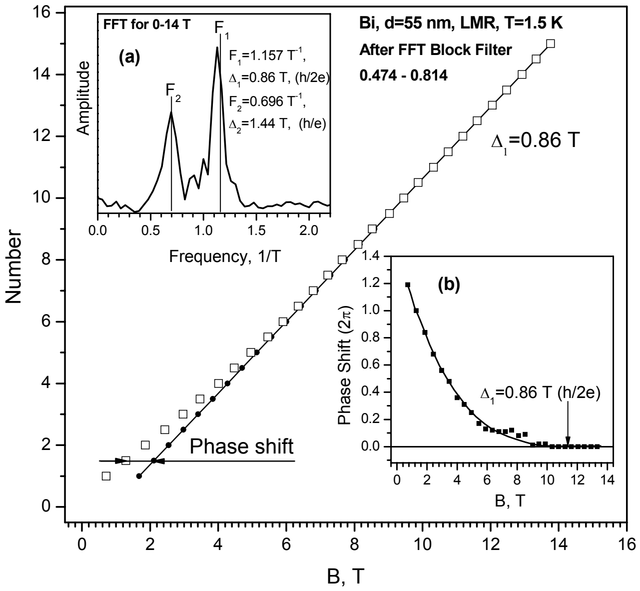

Fig. 5. Quantum number of the actual h/2e oscillation versus magnetic field for a 55 nm Bi nanowire. Insert (a): FFT spectra of the longitudinal MR oscillations; insert (b): Changes of extrema positions vs. B for h/2e oscillations which were converted into the values of phase shift of high field harmonic oscillation.

dependencies have "semiconductor" character, i.e. the resistance grows in the whole range of temperatures. For T>100 K, the nanowires' resistance $R(T)\sim\exp(\Delta/2k_BT)$. Δ is found to be 10±5 meV for both the 55 nm and 73 nm wires. Following Choi et al [28], Δ is interpreted as the energy gap between the electron and hole band in the core of the nanowires. The values of Δ that are observed are in rough agreement with theoretical work [2] which indicates that the band overlap decreases substantially below the value for bulk Bi (38 meV) because of quantum confinement. Therefore, one expects the electron and hole densities in our nanowires to be less than in bulk Bi. It can be surmised that the low-temperature electronic transport that is observed is mediated by surface states.

Insert (b) in Fig. 10 shows the oscillation part of magnetic field derivative of the longitudinal MR of 55 nm nanowire. The inset (a) in Fig. 11 shows Fast Fourier Transform (FFT) spectra of this oscillation. Longitudinal MR oscillations that are equidistant in the magnetic field and decrease in amplitude have been observed for the first time in magnetic fields up to 14 T in Bi single crystal nanowires d < 80 nm at T=1.4 ÷ 4.2 K [23, 24]. In contrast to oscillations that have been observed in thick Bi microwires (0.2<d<0.8 μm, $\Delta B_1=h/e$ and $\Delta B_2=1.4h/e$) [20, 21] they exist in wide range of magnetic field (up to 14 T) and have two periods: $\Delta B_1=h/e$ and $\Delta B_2=h/2e$. Finding the h/e and the $h/2e$ modes together is not surprising; this has been observed in rings where it has been interpreted to be caused by the interference of electrons that encircle the ring twice [29]. Due to SMSC in our nanowires carrier concentration in wires core very small, it's unlikely that they excite this oscillations. Angle-resolved photoemission spectroscopy (ARPES) studies of planar Bi surfaces have shown that they support surface states, with carrier densities Σ of around $5\text{x}10^{12}$ cm^{-2} and large effective masses m_Σ of around 0.3[30]. The observed effects are consistent with theories of the surface of nonmagnetic conductors whereby Rashba spin-orbit interaction gives rise to a significant population of surface carriers [25]. Measurements of the Fermi surface of small diameter bismuth nanowires employing the Shubnikov–de Haas method [31] have also been interpreted by assuming the presence of surface charge carriers with $\Sigma=2\text{x}10^{12}$ cm^{-2} and with the same effective mass as in ARPES.

Given the bulk electron n and hole p densities (in undoped Bi, $n=p=3\text{x}10^{17}$ cm^{-3} at 4 K) and the surface density Σ measured by ARPES or by SdH, one expects the surface carriers to become a clear majority in nanowires with diameters below 100 nm at low temperatures; the ratio of surface carrier density to bulk electrons or holes is 12 for 55 nm wires. At that point, the nanowire should effectively become a conducting tube. The electrical transport properties of nanotubes are unique because the wavelike nature of the charge carriers manifests itself as a periodic oscillation in the electrical resistance as a function of the enclosed magnetic flux $\Phi=(\pi/4)d^2B$.

Using FFT analysis we have separated every frequency for longitudinal MR of 55 nm nanowire. As it has appeared h/e oscillation is harmonic but the extrema of h/2e oscillation lie on a straight line (Fig. 5) only for B > 8T and step-by-step deviate from it at low magnetic fields. After converting the low field extrema positions to the phase shift of high field harmonic oscillations the phase shift curve (Fig. 5, insert (b)) was obtained. Mathematic simulation was used for testing of this method. From B ~ 8 T down to B=0 the extremums of h/2e oscillations are shifted up to 3π at B=0 which is the manifestation of Berry phase shift due to electron moving in nonuniform magnetic field.

Derivative of MR for 55 nm bismuth nanowire was measured at various inclined angels up to 90^o of magnetic fields relative to nanowire's axis in two mutually perpendicular planes. The observed angle variation of the periods are not in the agreement with theoretical dependence $\Delta(\alpha)=\Delta(0)/\cos\alpha$ of the size effect oscillations of the "flux quantization" type when period of oscillations in magnetic field depends only on the component B_X, parallel to the axis of the cylindrical sample. According to the experimental data the shifts of oscillation frequencies at the same angles depend on plane of sample rotation. Derivative of MR was measured at various rotational angles of nanowire when axis of the wire was mounted perpendicularly to the magnetic field. Even in this case the equidistant oscillations of MR exist in transverse magnetic fields under certain rotation angles. Moreover the SdH oscillation with period $\Delta(1/B)=0.06\ T^{-1}$ arise at the orientation magnetic field almost parallel to C_2 axis (and perpendicular to nanowire's axis) from carriers with effective masses of 0.25 m_0 which is close to the data obtained by ARPES of planar Bi surfaces for carriers on surface states. [30] This means that oscillations nature connected with Bi Fermi surfaces and very complicated. We connect the existence of h/2e oscillations with weak localizations on surface states of Bi nanowires according to the AAS theory.

ACKNOWLEDGMENTS

This work was supported by the Academy of Sciences of Moldova Grant No 09.808.05.02A.

REFERENCES

[1] L.D. Hicks, M.S. Dresselhaus, Phys. Rev. B **47**, 16631 (1993).

[2] Y. Lin, X. Sun, and M. S. Dresselhaus, Phys. Rev. B **62**, 4610 (2000).

[3] V. S. Edel'man, Adv. Phys. **25**, 555 (1976).

[4] G. E. Smith, G. A. Baraf, and J. M. Rowell, Phys. Rev. **135**, A1118 (1964).

[5] F. Y. Yang, et al., Phys. Rev. B **61**, 6631 (2000).

[6] Z. Zhang, X. Sun, M. S. Dresselhaus, J. Y. Ying, and J. P. Heremans, APL, **73**, 1589 (1998).

[7] Y. Aronov and D. Bohm, Phys. Rev. **115**, 485 (1959).

[8] I.O Kulik, Pis'ma Zh. Exp. Teor. Fiz. **5**, 423 (1967) [JETP Lett. **5**, 145 (1967)].

[9] A. Shablo, T. Narbut, S. Tyurin, and I. Dmitrenko, Pis'ma Zh. Exp. Teor. Fiz. **13**, 457 (1974) [JETP Lett. **19**, 146 (1974)]

[10] R. Dingle, Proc. Roy. Soc. London, Ser. A 212, **47** (1952).

[11] B.L.Al'tshuler, A.G.Aronov, and B.Z.Spivak, Zh. Exp. Teor. Fiz., Pis. Red., **33**, 101 (1981) [JETP Lett. **33**, 94 (1981)].

[12] D.Yu.Sharvin, and Yu.V.Sharvin, Zh. Exp. Teor. Fiz., Pis. Red., **34**, 285 (1981) [JETP Lett., **34**, 272 (1981)].

[13] V. Chandrasekhar, M. J. Rooks, S. Wind, and D. E. Prober, Phys. Lett. **55** 1610 (1985)

[14] A. E. Hansen, A. Kristensen, S. Pedersen, C. B. Sorensen, and P. E. Lindelof, Phys. Rev. B **64**, 045327 (2001).

[15] J. B. Yau, E. P. De Poortere, and M. Shayegan, Phys. Rev. Lett. **88**, 146801 (2002).

[16] M. Konig, A. Tschetschetkin, E. M. Hankiewicz, J. Sinova, V. Hock, V. Daumer, M. Schafer, C. R. Becker, H. Buhmann, and L. W. Molenkamp, Phys. Rev. Lett. **96**, 076804 (2006).

[17] A.G. Aronov and Y.V.Sharvin, Rev. Mod. Phys. **59**, 755 (1987).

[18] A. Bachtold, A. Strunk, J-P Salvetat, J-M Bonard, L. Forro, T. Nussbaumer, and C. Schonenberger, Nature **397**, 673 (1999).

[19] T.E.Huber, K.Celestine, and M.J.Graf, Phys. Rev. B, **67**, 245317 (2003).

[20] N.B. Brandt, D.V. Gitsu, A.A. Nikolaeva, and Ya.G. Ponomarev, Zh. Exp. Teor. Fiz., **72**, 2332-2334, (1977) [Sov. Phys. JETP, **45**, 1226 (1977)].

[21] N.B. Brandt, E.N.Bogachek, D.V. Gitsu, G.A.Gogadze, I.O.Kulik, A.A. Nikolaeva, and Ya.G. Ponomarev, Fiz. Nizk. Temp., **8**, 718 (1982) [Sov. J. Low Temp. Phys. **8**, 358 (1982)]

[22] E. N. Bogachek and G. A. Gogadze, Zh. Eksp. Teor. Fiz. **63**, 1839 (1972) [Sov. Phys. JETP **36**, 973 (1973)].

[23] D. Gitsu, T. Huber, L. Konopko, and A. Nikolaeva, AIP Conference Proceedings **850**, p. 1409 (2006).

[24] A. Nikolaeva, D. Gitsu, L. Konopko, M.J. Graf, and T. E. Huber, Phys. Rev. B, **77**, 075332 (2008).

[25] D. Gitsu, L. Konopko, A. Nikolaeva, and T. E. Huber, Appl. Phys. Lett. **86**, 102105 (2005).

[26] J. Heremans, C. M. Thrush, Y.-M. Lin, S. Cronin, Z. Zhang, M. S. Dresselhaus, and J. F. Mansfield, Phys. Rev. B **61**, 2921 (2000).

[27] R. G. Chambers, Proc. Roy. Soc. A **202**, 378 (1950).

[28] D. S. Choi, A. A. Balandin, M. S. Leung, G. W. Stupian, N. Presser, S. W. Chung, J. R. Heath, A. Khitun, and K. L. Wang, Appl. Phys. Lett. **89**, 141503 (2006).

[29] V. Chandrasekhar, M. J. Rooks, S. Wind, and D. E. Prober, Phys. Lett. **55** 1610 (1985)

[30] Yu. M. Koroteev, G. Bihlmayer, J. E. Gayone, E. V. Chulkov, S. Blugel, P. M. Echenique, and Ph. Hofmann, Phys. Rev. Lett. **93**, 046403 (2004).

[31] T. E. Huber, A. Nikolaeva, D. Gitsu, L. Konopko, C. A. Foss, Jr., and M. J. Graf, Appl. Phys. Lett. **84**, 1326 (2004).