Continuous spin reorientation transition in epitaxially grown antiferromagnetic NiO thin films

J. Li,[1] E. Arenholz,[2] Y. Meng,[1,3] A. Tan,[1] J. Park, [1] E. Jin,[1] H. Son,[1] J. Wu,[1,4] C. A. Jenkins,[2] A. Scholl,[2] Chanyong Hwang,[5] and Z. Q. Qiu[1]

[1] *Department of Physics, University of California at Berkeley, Berkeley, California 94720, USA*

[2] *Advanced Light Source, Lawrence Berkeley National Laboratory, Berkeley, California 94720, USA*

[3] *Institute of Physics, Chinese Academy of Science, Beijing, P. R. China*

[4] *Brookhaven National Laboratory, Upton, NY 11973*

[5] *Korea Research Institute of Standards and Science, Yuseong, Daejeon 305-340, Korea*

Abstract

Fe/NiO/MgO/Ag(001) films were grown epitaxially and the Fe and NiO spin orientations were determined using X-ray Magnetic Dichroism. We find that the NiO spins are aligned perpendicularly to the Fe spins. Analyzing both the in-plane and out-of-plane spin components of the NiO layer, we demonstrate unambiguously that the NiO spins undergo a continuous spin reorientation transition from the in-plane to out-of-plane directions when tuning the in plane epitaxial strain from compressive to tensile with increasing the MgO thickness.

PACS numbers: 75.70.Ak

Controlling the spin direction of a magnetic nanostructure is one of the most important tasks in spintronics research [1]. In particular, it has been an outstanding topic for a long time to tailor the spin direction from the in-plane to out-of-plane directions of a magnetic thin film — the so-called spin reorientation transition (SRT). For ferromagnetic (FM) thin films, the significance of the SRT has been well recognized because of its crucial role in both technological applications (e.g., longitudinal and perpendicular magnetic recording) [2] and in fundamental understanding of two-dimensional magnetism (e.g., magnetic stripe [3,4,5] and bubble domain phases [6,7]). For antiferromagnetic (AFM) thin films, however, the SRT has not yet been realized despite of its importance. Different from the FM thin films where the SRT is generally realized by the competition between the surface perpendicular magnetocrystalline anisotropy and the volume in-plane shape anisotropy, a realization of the SRT in an AFM thin film depends entirely on a fine tuning of the magnetocrystalline anisotropy across zero because of the absence of magnetic shape anisotropy in AFM thin films. To date, the most promising method of tuning the magnetocrystalline anisotropy of an AFM thin film is to induce lattice distortions either in the multiferroic compounds [8,9] or by growing the AFM thin film on a lattice mismatched substrate [10]. The multiferroics research employs the coupling of ferroelectric and magnetic polarization vectors and faces various challenges such as the leakage current and repeatable switching. The lattice mismatching method employs a fine tuning of the AFM strain to modify the magnetocrystalline anisotropy of the AFM thin film [11]. A promising system in the latter approach are epitaxial NiO/Ag and NiO/MgO thin films as NiO grows compressed on Ag(001) and in tensile strain on MgO (001) (a_{Ag} =4.086Å < a_{NiO} =4.176Å < a_{MgO} =4.212Å) [15]. It was shown that epitaxial NiO films prefer an in-plane spin direction near the Ag interface and an out-of-plane spin direction near the MgO interface [12]. However, it is not clear if the NiO spins in the MgO/NiO(30ML)/Ag(001) indeed could break into two regions with different spin directions. Recently, it was shown that a FM overlayer can align the NiO spins in the surface near region of a NiO single crystal into the surface plane [13,14]. Moreover, FM hysteresis loop measurements of the Fe layer support the fact that the NiO spins have an in-plane direction in Fe/NiO/Ag(001) and an out-of-plane direction in Fe/NiO/MgO(001) [15]. Despite this progress, a continuous SRT from in-plane to out-of-plane directions has never been realized in an AFM thin film. In this Letter, we report a study of the epitaxially grown Fe/NiO/MgO/Ag(001) system. We demonstrate for the first

time that the NiO spins in this system undergo a continuous SRT from in-plane to out-of-plane directions with varying epitaxial strain.

Fe/NiO/MgO/Ag(001) films were grown epitaxially onto a Ag(001) substrate at room temperature. While the Fe and MgO films were grown by evaporating Fe and MgO from two e-beam evaporators, the NiO film was prepared by growing Ni at an oxygen pressure of ~$1x10^{-6}$ Torr. The formation of single crystalline Fe/NiO/MgO/Ag(001) film was confirmed by Low Energy Electron Diffraction (LEED) with the crystal relation of Fe[100]//NiO[110]//MgO[110]//Ag[110]. The Fe and NiO thicknesses were fixed and the MgO film was grown into a wedge shape (0-60ML over 6mm). The varying MgO layer thickness leads to a continuous change in its in-plane lattice constant from that of Ag (4.09Å) towards that of MgO (4.21Å) to modulate the strain of the epitaxial NiO overlayer. The sample of Fe(10ML)/NiO(25ML)/MgO(wedge)/Ag(001) was covered by a 10ML Ag protection layer and brought to beamline 4.0.2 of the Advanced Light Source for X-ray magentic circular dichroism (XMCD) and X-ray magentic linear dichroism (XMLD) [16] measurements. The thickness-dependent measurement was obtained by moving the sample along the MgO wedge direction. The x-ray beam size in the wedge direction is 100μm which leads to a thickness variation of 1ML within the x-ray spot.

We first discuss the XMLD result at normal incidence of the x-rays with a linear polarization. An in-plane magnetic field was applied to align the Fe magnetization along the NiO[010] axis and obtain a single domain configuration in the entire system. The X-ray Absorption Spectrum (XAS) of the Ni L_2 edge was taken at different polarization angles [Fig. 1(a)]. At 0ML MgO thickness (d_{MgO}=0ML), the XAS at polarization $\phi=0^o$ and $\phi=90^o$ are different [Fig. 1(b)], showing an in-plane NiO spin component at d_{MgO}=0ML. In contrast, the XAS at $\phi=0^o$ is the same as the XAS at $\phi=90^o$ in the d_{MgO}=35ML sample [Fig. 1(b)], showing an absence of the in-plane NiO spin component at d_{MgO}=35ML. Fe magnetic hysteresis loop measurements were obtained using XMCD from the Fe edge in grazing incidence geometry where only the in-plane Fe magnetization is detected. The result [Fig. 1(b)] shows that the d_{MgO}=0ML sample has a much greater coercivity (H_C=346 Oe) than the d_{MgO}=35ML sample (H_C=90 Oe). Taking into account that only in-plane NiO spins enhance the in-plane Fe coercivity, the Fe hysteresis loop supports the XMLD result that the NiO spins should have an in-plane direction in the d_{MgO}=0ML sample and a zero in-plane component in the d_{MgO}=35ML sample. To identify the in-plane NiO spin direction, we measured the L_2 ratio

(R_2), which is defined as the ratio of the lower energy peak divided by the higher energy peak in the XAS, as a function of ϕ. The result [Fig. 1 (c)] shows that R_2 follows the relation of $R_2(\phi) = A\cos^2\phi + B$ with the minimum at ϕ=0° (NiO[100]). Then from the established XMLD result [17], the in-plane NiO spin axis must be parallel to the NiO[100] axis. We then conclude that the Fe and NiO in-plane spins are coupled perpendicularly as shown in Fig. 1(a). Recalling that the L_2 ratio difference $\Delta R_2 \equiv \left|R_2(0^0) - R_2(90^0)\right|$ is proportional to S_x^2 of the NiO spins, the result of Fig. 1(c) implies a continuous SRT of the NiO spins from the in-plane (x-axis) to out-of-plane (z-axis) directions within the x-z plane.

Since the XMLD at normal incidence of the x-rays is only sensitive to the in-plane spin component of the NiO spins, we further performed XMLD measurement at off-normal incidence [Fig. 2(a)] of the x-rays with the s-polarization measurement ($\vec{E} // x$) to pick up the x-component of the NiO spins and the p-polarization measurement ($\vec{E}$ within the yz-plane) to pick up the z-component of the NiO spins. We chose θ=20° to maximize the z-projection of the p-polarization vector without spreading too much of the x-ray beam on the sample surface. We first measured the XAS by aligning the Fe magnetization to the y-axis (e.g., NiO in-plane spins parallel to the x-axis). For d_{MgO}=0ML sample, the XAS at s-polarization has a lower L_2 ratio than at s-polarization [Fig. 2(b)] which is consistent with the result of $\vec{S}_{NiO} // x$. For d_{MgO}=35ML sample, the XAS at s-polarization has a greater L_2 ratio than at p-polarization [Fig. 2(b)], confirming the existence of a z-component NiO spins, i.e., the absence of the in-plane NiO spin component for the d_{MgO}=35ML sample shown in Fig. 1(b) is due to the perpendicular alignment of the NiO spins (z-axis). Therefore the results of Fig. 1 and 2 together demonstrate the existence of a SRT of the NiO spins from in the sample surface plane to perpendicular to the sample surface (in the plane perpendicular to the Fe spin axis) as the d_{MgO} changes from d_{MgO}=0ML to d_{MgO}=35ML. To ensure that the result of Fig. 2 is of magnetic origin rather than crystal field effect [18], we did the measurement by fixing the x-ray polarization at the s-polarization mode ($\vec{E} // x$) but to change the Fe spin from the NiO[010] to the NiO [100] directions [Fig. 3(a)]. For d_{MgO}=0ML sample, the XAS shows a lower L_2 ratio for $\vec{S}_{Fe} // y$ than for $\vec{S}_{Fe} // x$ [Fig. 3(b)], consistent with an in-plane direction of the NiO spins which are coupled perpendicular to the Fe spins. For d_{MgO}=35ML sample the XAS shows identical spectrum for $\vec{S}_{Fe} // x$ and $\vec{S}_{Fe} // y$ [Fig. 3(b)], consistent with a

perpendicular direction of the NiO spins ($\vec{S}_{NiO} // z$).

Next, the L_2 ratio was measured along the MgO wedge for both s-polarized [$R_2^s(d_{MgO})$] and p-polarized [$R_2^p(d_{MgO})$] x-rays by aligning the Fe magnetization in the y-axis with an external magnetic field. Since under this condition the R_2^s is proportional to the projection of NiO spin in the x-axis (e.g. $R_2^s = AS_{x,NiO}^2 + B$) and the R_2^p is proportional to the projection of NiO spin in the z-axis (e.g. $R_2^p = A'S_{z,NiO}^2 + B'$) [19], the quantities of $r_2^s \equiv \frac{R_2^s(d_{MgO}) - R_2^s(\infty)}{R_2^s(0) - R_2^s(\infty)}$ and $r_2^p \equiv \frac{R_2^p(d_{MgO}) - R_2^p(0)}{R_2^p(\infty) - R_2^p(0)}$ represent approximately the quantities of $\frac{S_{x,NiO}^2(d_{MgO}) - S_{x,NiO}^2(\infty)}{S_{NiO}^2} = \frac{S_{x,NiO}^2(d_{MgO})}{S_{NiO}^2}$ and $\frac{S_{z,NiO}^2(d_{MgO}) - S_{z,NiO}^2(0)}{S_{NiO}^2} = \frac{S_{z,NiO}^2(d_{MgO})}{S_{NiO}^2}$, respectively. Taking $R_2^{s,p}(0)$ from the d_{MgO}=0ML sample and $R_2^{s,p}(\infty)$ from the d_{MgO}=45ML sample region, we obtained r_2^s and r_2^p as a function of d_{MgO}. The result (Fig. 4) shows that both r_2^s and r_2^p change monotonically with d_{MgO} but the sum of r_2^s and r_2^p always remains unity ($r_2^s + r_2^p = 1$). This result proves that the NiO spins undergo a continuous SRT in the xz plane with increasing d_{MgO}. The Fe coercivity was also measured and the monotonic decrease of the Fe coercivity (Fig. 4) supports the conclusion that the NiO spins undergo a continuous SRT as a function of d_{MgO}. It should be mentioned that we also studied the Fe/NiO/Ag(wedge)/MgO(001) system and the result does not show a clear continuous SRT of the NiO spins. This might be explained by the details in growth of Ag on MgO as compared to MgO on Ag which lead to a complex dependence of the Ag in-plane lattice constant on the Ag layer thickness [20]. Further detailed studies on how the spacer layer affects the strain and magnetic anisotropy of the NiO film should be performed in the future.

In summary, we studied epitaxially grown Fe/NiO/MgO(wedge)/Ag(001) by XMLD and magnetic hysteresis loop measurements. We show unambiguously that the NiO spins undergo a continuous SRT from in-plane to out-of-plane directions as function of in-plane epitaxial strain.

Acknowledgement

This work was supported by the National Science Foundation under DMR-0803305,

by KICOS through Global Research Laboratory project, and by the Chinese Education Department (JSP, YM, JL, EJ, JS, AT, JW, CH, HWZ, ZQQ). ALS experimental work was supported by the U.S. Department of Energy under Contract No. DE-AC02-05CH11231 (EA, KJ, AS).

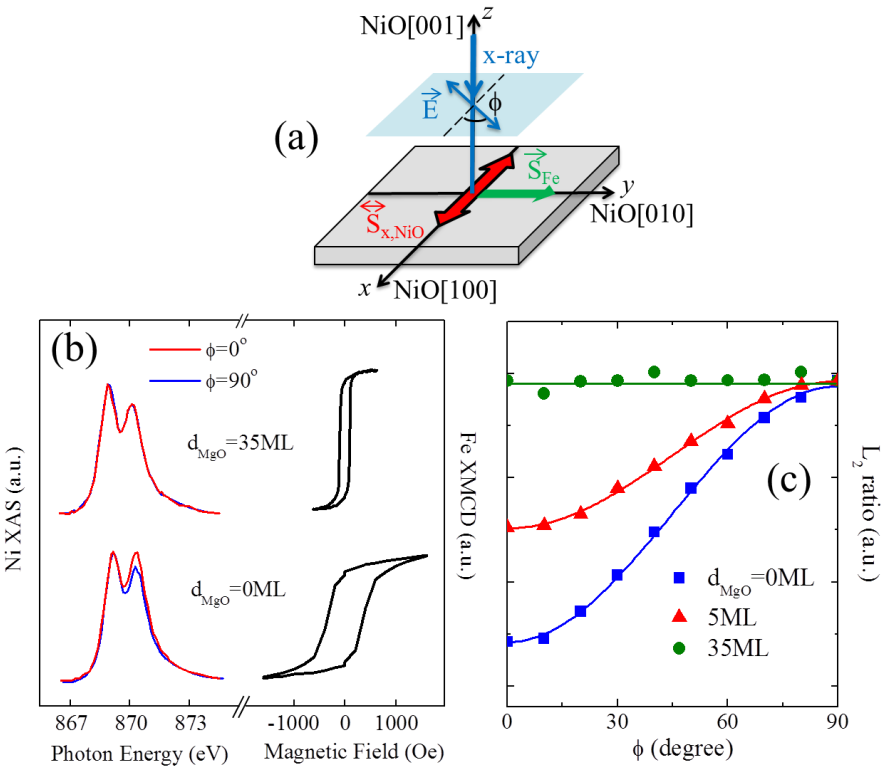

Fig. 1: (color online) (a) Schematic drawing of the measurement condition. (b) XAS from the Ni L2 edge and Fe magnetic hysteresis loops from the Fe XMCD measurement. (c) L_2 ratio versus the polarization angle. The result shows that the NiO in-plane spin component decreases to zero as the MgO thickness increases to 35ML.

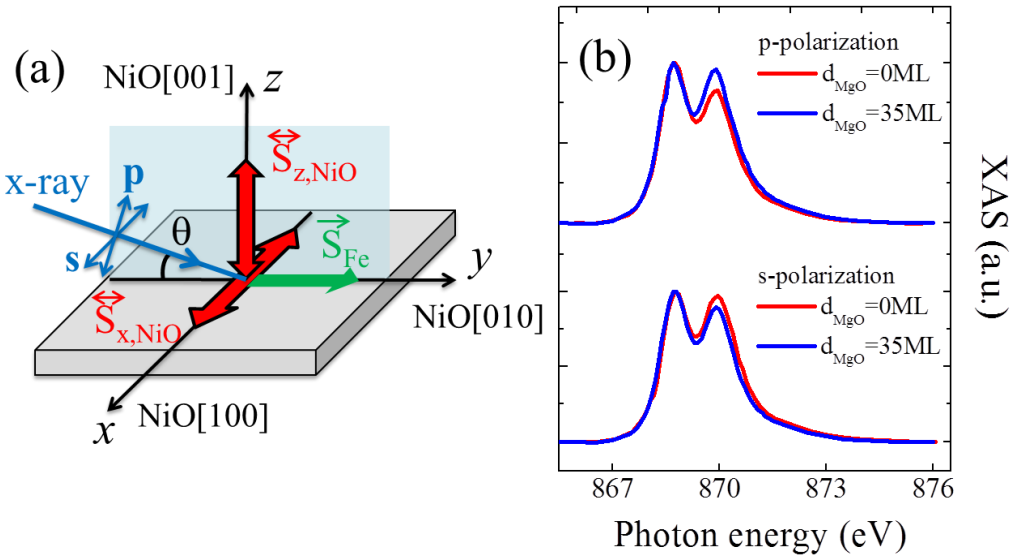

Fig. 2: (color online) (a) Schematic drawing of the measurement condition. (b) Ni XAS for d_{MgO}=0ML and d_{MgO}=35ML samples. The opposite behaviors for s- and p-polarized x-rays suggests that the NiO spins undergo a SRT from the in-plane to out-of-plane directions as d_{MgO} increases from 0 to 35 ML.

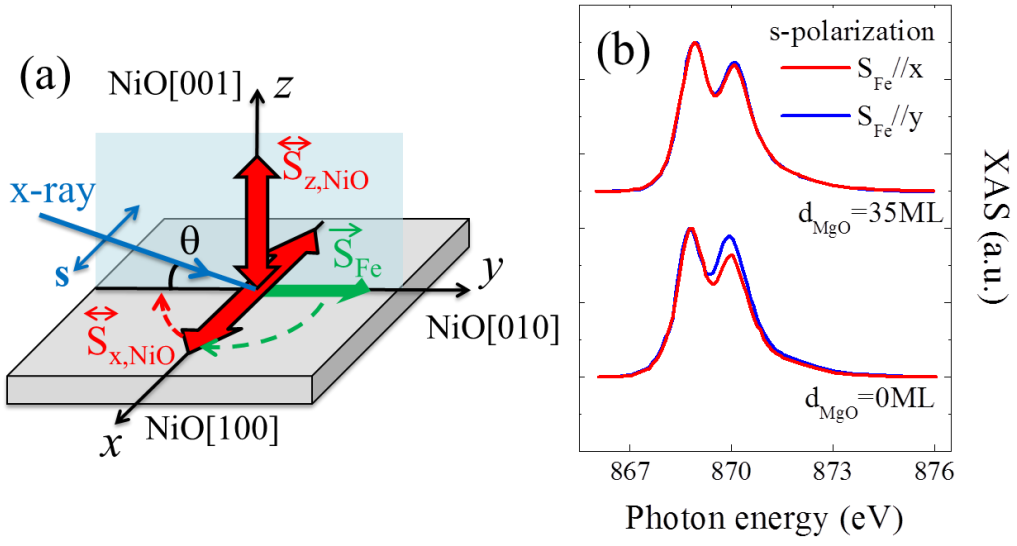

Fig. 3: (color online) (a) Schematic drawing of the measurement condition. (b) Ni XAS for $\vec{S}_{Fe} // x$ and $\vec{S}_{Fe} // y$. The identical XAS for $\vec{S}_{Fe} // x$ and $\vec{S}_{Fe} // y$ from the d_{MgO}=35ML sample show that the NiO spins are perpendicular to the sample surface for d_{MgO}=35ML.

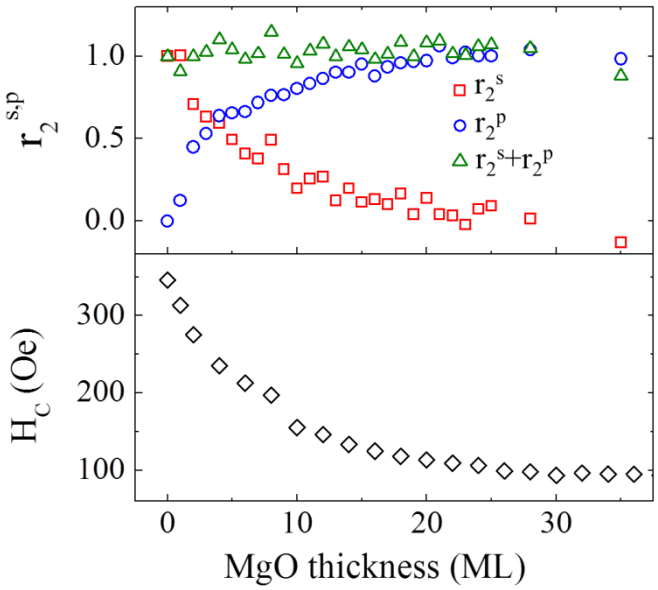

Fig. 4: (color online) The relative L_2 ratio r_2^s and r_2^p for s- and p-polarizations (see text) under the measurement condition shown in Fig. 2, and the Fe coercivity as a function of d_{MgO}. r_2^s and r_2^p represent $\vec{S}_{x,NiO}^2/\vec{S}_{NiO}^2$ and $\vec{S}_{z,NiO}^2/\vec{S}_{NiO}^2$, respectively. The monotonic change of r_2^s from 1 to 0 (r_2^p from 0 to 1) and the fact of $r_2^s + r_2^p = 1$ show that the NiO spins undergo a continuous SRT from the in-plane to out-of-plane directions within the xz plane as d_{MgO} increases from 0 to ~30ML.

References:

1. S.D. Bader and S.S.P. Parkin, Annual Review of Condensed Matter Physics, **1**: 71 (2010).
2. Mitsumasa Oshiki**,** J. Magn. Magn. Mat., in press.
3. D. P. Pappas, K.–P. Kämper, and H. Hopster, Phys. Rev. Lett. **64**, 3179 (1990).
4. R. Allenspach and A. Bischof, Phys. Rev. Lett. **69**, 3385 (1992).
5. Y. Z. Wu, C. Won, A. Scholl, A. Doran, H. W. Zhao, X. F. Jin, and Z. Q. Qiu, Phys. Rev. Lett. **93**, 117205 (2004).
6. J. Choi, J. Wu, C. Won, Y. Z. Wu, A. Scholl, A. Doran, T. Owens, and Z. Q. Qiu, Phys. Rev. Lett. **98**, 207205 (2007).
7. N. Saratz, A. Lichtenberger, O. Portmann, U. Ramsperger, A. Vindgni, and D. Pescia, Phys. Rev. Lett. **104**, 077203 (2010).
8. N. A. Spaldin, S.-W. Cheong and R. Ramesh, Physics Today **63** (10), 38 (2010).
9. and, Nature Materials **6**, 21 (2007).
10. A. Boussendel, N. Baadji, A. Haroun, H. Dreyssé, and M. Alouani, Phys. Rev. B **81**, 184432 (2010).
11. Suman Mandal, Krishnakumar S. R. Menon, Francesco Maccherozzi, and Rachid Belkhou Phys. Rev. B **80**, 184408 (2009).
12. S. Altieri, M. Finazzi, H. H. Hsieh, H.-J. Lin, C. T. Chen, T. Hibma, S. Valeri, and G. A. Sawatzky, Phys. Rev. Lett. **91**, 137201 (2003).
13. H. Ohldag, A. Scholl, F. Nolting, S. Anders, F. U. Hillebrecht, and J. Stöhr, Phys. Rev. Lett. **86**, 2878 (2001).
14. H. Ohldag, G. van der Laan, and E. Arenholz, Phys. Rev. B **79**, 052403 (2009).
15. Wondong Kim, E. Jin, J. Wu, J. Park, E. Arenholz, A. Scholl, Chanyong Hwang, and Z. Q. Qiu, Phys. Rev. B **81**, 174416 (2010).
16. B. T. Thole, G. van der Laan, and G. A. Sawatzky, Phys. Rev. Lett. **55**, 2086 (1985).
17. Elke Arenholz, Gerrit van der Laan,, Rajesh V. Chopdekar, and Yuri Suzuki, Phys. Rev. Lett. 98, 197201 (2007).
18. M. W. Haverkort, S. I. Csiszar, Z. Hu, S. Altieri, A. Tanaka, H. H. Hsieh, H.-J. Lin, C. T. Chen, T. Hibma, and L. H. Tjeng, Phys. Rev. B **69**, 020408 (2004).
19. Precisely speaking, this is true only for x-ray polarization along certain crystal axis [e.g., NiO(100)]. Thus the $r_2^p = S_{z,NiO}^2(d_{MgO})/S_{NiO}^2$ should be an approximation in the small θ limit. See Ref. 10 for details.
20. O. Robach, G. Renaud, and A. Barbier, Phys. Rev. B **60**, 5858 (1999).