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Dramatic depression of Curie temperature for magnetic Co/Cu(1 0 0) ultrathin films upon deposition at elevated temperature

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Abstract

The correlation between the crystalline structures and magnetic properties, such as magnetization and Curie temperature (T_C) were investigated systematically by varying the deposition temperature of 2 ML fcc cobalt films on Cu(1 0 0) substrate. At variation of the deposition temperature a dramatic change in Curie temperature and coercivity of the films was observed. The drastic raising of Curie temperature were found for the films with the deposition temperatures from 340 K ($T_C = 170$ K) down to 275 K ($T_C = 325$ K). A simple theoretical estimation was proposed to evaluate the Curie temperature of the films in terms of magnetic moment as well as anisotropy with quantitative success. The enhanced Curie temperature can be attributed to the increase of the magnetic moments due to the alteration of film morphology with various deposition temperatures.

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1. Introduction

Magnetic ultrathin films have attracted a lot of interest in the past decades due to their unique properties different from those in the bulk. The magnetic properties in these systems are exceedingly sensitive to the crystalline structure as well as morphology, which can be altered by growth conditions, such as film coverage and deposition temperature. It has been demonstrated the magnetic phase transition is affected by the coverage as well as deposition temperature of the Fe/Cu(1 0 0) films [1–5]. The dependence of the magnetic phase of fcc-Fe on the lattice constant (3.59 Å for AFM [6] and 3.64 Å for FM phase [7]) was shown to be the origin of this behavior [8]. Similarly, the influence of deposition temperature on SRT was also observed in the Ni/Cu(1 0 0) systems [9]. The critical thickness for the low-temperature deposited film was found to be about 1 monolayer (ML) less than that for the high-temperature deposited film. This observation was attributed to the reduced surface anisotropy in low-temperature deposited films caused by the increase of the number of step

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edge atoms. There also exists the deposition-temperature dependent behavior in Co/Cu(1 0 0). The coercivity was found to be sensitive to the deposition temperature and annealing history of the films [10–12]. The morphology of the films plays an important role to interpret these observations.

Further, the characteristically thermodynamic property for the ultrathin films is the reduced Curie temperature (T_C) owing to finite size scaling effect [8,13–15]. The previous studies for Fe/Cu_3Au(1 0 0) [16] and Co/Cu(1 0 0) [10–12] also suggest the variations of T_C for the films at different deposition temperature. Nevertheless, a systematic study is still lacking to clarify the influence of the deposition temperature on Curie temperature. For better understanding of the correlation between the Curie temperature and the deposition temperature of the ultrathin films, a systematic investigation is of the utmost importance. In addition, there exists in-plane magnetization only in Co/Cu(1 0 0) films for all coverage of Co [13]. Therefore it is a good candidate for studying the phase transition between the ferromagnetic and paramagnetic phase, that is, the Curie temperature of the ultrathin films without the ambiguity cause by the spin-reorientation transition. In this article, we report on the significant change in Curie temperature of the Co/Cu(1 0 0) ultrathin films at variation of the deposition temperature. The analyses of the structure were performed in situ to provide a quasi-simultaneous structural information with the magnetic measurements. The Curie temperature was found to be drastically enhanced while lowering the deposition temperature below 250 K. We also proposed a simple theoretical estimation to evaluate T_C of the films deposited at different temperatures. It will be shown that the enhanced Curie temperature is accompanied by the increase of remanent Kerr signal, which may be due to the change of the number of the island edge atom at the alteration of morphology.

2. Experiment

The experiments were carried out in an ultrahigh vacuum chamber with the base pressure of 5×10^{-10} mbar. Auger electron spectroscopy (AES), low-energy electron diffraction (LEED), medium-energy electron diffraction (MEED), magneto-optical Kerr effect (MOKE), ion beam sputter, and molecular beam epitaxy evaporators were installed in this chamber for the in situ study. The Cu(1 0 0) substrate was cleaned by cycling processes of 2-KeV argon ion bombardment and annealing to 800 K for 5 min. The crystallographic ordering and cleanness of the surface were checked by LEED and AES, respectively. The background pressure during cobalt evaporation was kept at 1×10^{-9} mbar under the deposition rate of 0.5 ML per min. The layer-by-layer growth of cobalt films was simultaneously monitored by MEED during the thermal evaporation process such that the coverage of the ultrathin films can be precisely controlled in the accuracy of 0.05 ML. The thickness of films was also confirmed by means of AES. After the film being deposited, the crystalline structure and interlayer distance of the surface were measured at the temperature of 200 K via LEED and LEED $I(E)$ in the kinematic approximation [17,18], respectively. The MOKE measurement comprises He–Ne lasers ($\lambda = 632.8$ nm) as the light sources and a photodiode as the light detector. In addition, the photoelastic modulator combined with the Lock-in technique were applied in our measurements for the acquisition of signals in a good signal to noise ratio. The magnetic hysteresis loops were taken by means of the longitudinal MOKE which is normalized by reflected beam intensity to ensure the magnitude of Kerr intensity can be compared for various measurements. The remanent Kerr signal versus temperature was used to determine the Curie temperature of the films.

3. Results

As introduced, the Curie temperature is sensitively affected by the coverage of the films [8,13–15] owing to the finite size scaling effect. For Co/Cu(1 0 0), T_C decreases linearly with thickness from 580 K for 3 ML to 120 K for 1.5 ML [13,19]. Thus, the determination of the film coverage plays an important role in our experiments. By well controlling deposition rate associated with the MEED

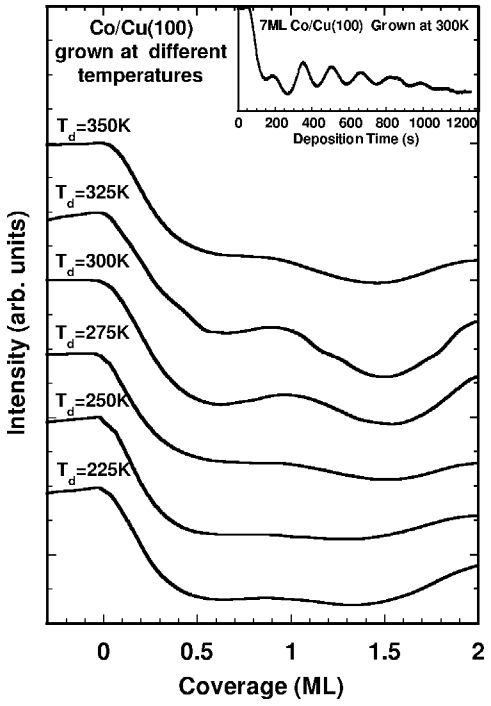

Fig. 1. MEED intensity oscillations for 2 ML Co/Cu(1 0 0) for various deposition temperatures. The inset shows that the MEED intensity oscillates with the coverage of Co up to 7 ML.

intensity monitoring shown in Fig. 1, the films can be prepared with accuracy of the coverage less than 0.05 ML [20]. Fig. 1 shows the MEED intensity oscillations for 2 ML Co/Cu(1 0 0) grown at different temperatures. Also, the MEED intensity of Co/Cu(1 0 0) oscillates up to 7 ML was shown in the inset. One can see that the amplitude of the first peak is less than that of the second peak for the reason that Co grows in partial two-layer-thick islands for the coverage below 2 ML to form the first layer [21,22]. Moreover, the equal spacing oscillations shown in the inset of Fig. 1 point out the deposition rate in our experiments is well controlled. Obviously the first peak of MEED intensity is more suppressed for the films grown at lower temperature. The previous study has shown that the damping diffraction intensity oscillation means pronounced roughness of the film [23]. Thus, our results imply that the morphology of the films deposited at lower temperature are rougher than that at the higher temperature, as the previous observation indicated [10,24]. This tendency can also be observed in the LEED pattern, in which the LEED spots for the films deposited at lower temperature were broader than those at higher deposition temperature.

The hysteresis loops of the Co films were measured in situ by longitudinal MOKE at various temperatures after the film deposited. The hysteresis loops of 2 ML Co grown upon Cu(1 0 0) at 325 and 225 K for different measurement temperature are shown in the left and right column of Fig. 2, respectively. It is manifest that the shape of the loops varies gradually from the square loop at low temperature to the flat curve at high temperature at which ferromagnetic ordering is broken.

The Curie temperature for the films was determined by sequentially taking hysteresis loops and measuring the remanent Kerr signals as a function of temperature. As depicted in Fig. 4 (the solid circles and curve), the Curie temperature of the 2 ML Co keeps almost invariant for the deposition temperatures (T_d) from 125 to 250 K but drops rapidly as T_d higher than 275 K. For $T_d = 340$ K, it is clear that T_C reaches an amazing value 170 K, which is about half of that (325 K) for $T_d = 125$ K. Furthermore, the critical exponent β [$M/M_s \sim (1 - T/T_C)^{\beta}$] was also estimated by fitting the curves for magnetization as a function of temperature, as shown in Fig. 3 ($T_d = 325$ K). The average value of β for the films deposited at different

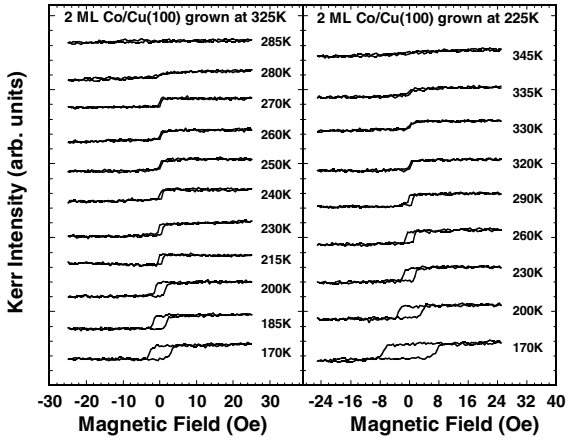

Fig. 2. Magnetic hysteresis loops for the 2 ML Co/Cu(1 0 0) films deposited at 325 K (left) and 225 K (right) at various measurement temperatures.

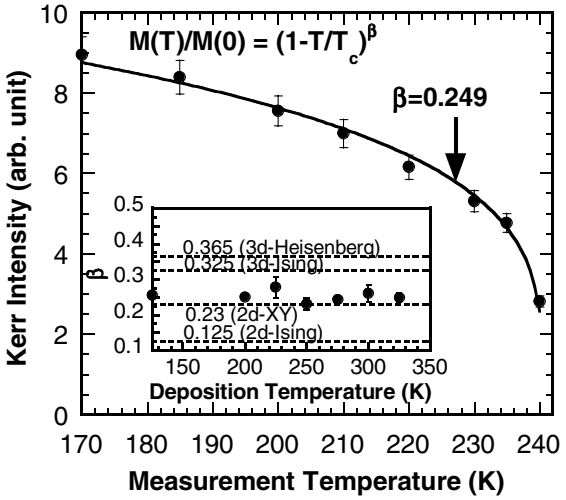

Fig. 3. The Kerr intensity versus measurement temperature for the film deposited at 325 K. The solid curve is the curve fitting of critical exponent β. Inset: critical exponent β of the 2 ML Co/Cu(1 0 0) films for different deposition temperatures (T_d). The dotted lines stand for the value expected by the theoretical models.

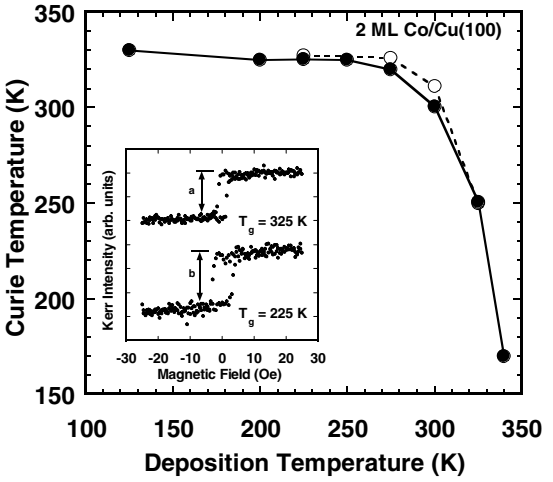

Fig. 4. Curie temperature (T_C) of the 2 ML Co/Cu(1 0 0) films for different deposition temperatures (T_d). The solid circles are the results in the experiments, the hollow ones are the simulated results from the remanent magnetization and coercive field taken at 200 K, as described in text. The hysteresis loops measured at 200 K for the films grown at 325 and 225 K are shown in the inset. Note that the ratio of remanent Kerr signal for 325 K (a) to that for 225 K (b) is $a : b = 1 : 1.1$.

temperatures is about 0.24 which lies close to the value expected by two-dimensional xy model [25] ($\beta \approx 0.23$), as shown in the in set of Fig. 3. It implies that the 2 ML-Co/Cu(1 0 0) films reveal the near two-dimensional magnetic behavior with fourfold symmetry. Our results for hysteresis loops with various in-plane angles also show the existence of biaxial anisotropy in these systems. In addition, there is no significant variance (excepted for the $T_d = 275$ K case with the larger error bar) for all films with different deposition temperatures. This suggests that the enhancement of Curie temperature is not intrinsically correlated to the critical exponent β of the film.

4. Theoretical estimation of T_C

The remanent Kerr signal and coercivity for the film deposited at 225 K are greater than those at 325 K, as shown in Fig. 2. It implies that the variation of Curie temperature may be attributed to the deviation of remanence as well as coercivity. The Curie temperature has been shown to be proportional to the square of the magnetic moment in the films [26,27]. By comparing the hysteresis loops measured at 200 K, one can find that the Kerr remanent signal (the same as the saturation one in these cases), which is proportional to the magnetization density, for $T_d = 225$ K is about 1.1 times of that for $T_d = 325$ K, as shown in the inset of Fig. 4. It can therefore induce about 20% enhancement of Curie temperature. It turns out that the enhancement of magnetization density for the low-temperature deposited films is the main contribution to the raising of the Curie temperature.

However, there is still a little deviation from the raising of Curie temperature taken in our study. In comparison with the films deposited at 225 K, the T_C enhancement for the 325 K-deposited films is about 9% deviation from the value estimated by considering the enhanced Kerr signals only. In addition to magnetization density, the Curie temperature depends on magnetic anisotropy as well. From the theory of the transition temperature of ultrathin films by means of renormalization group analysis [28], the two-dimensional transition temperature is shown to be proportional to

$[\ln(\pi^2 J/K)]^{-1}$, where J and K are the coupling constant in Heisenberg model and anisotropy constant, respectively. If one attributes this deviation to the difference of the anisotropies between these two films, the anisotropy of the film deposited at 225 K can be estimated to be 2.47 times of that at 325 K (for $K/J \sim 10^{-3}$). Surprisingly, it agrees well with the difference of the coercivity taken in the hysteresis loops for the films at $T_d = 325$ and 225 K. From the energy point of view, the film with the larger anisotropy energy is more difficult to reverse its magnetization by applied field and therefore reveals the larger coercivity. Thus, the 9% deviation may be corrected by taking the anisotropy into account. The relation of different Curie temperatures $T_{C,1}$ and $T_{C,2}$ can be therefore expressed as

$$\frac{T_{C,1}}{T_{C,2}} = \frac{M_1^2}{M_2^2} \times \frac{\ln[\pi^2 J/K_2]}{\ln[\pi^2 J/K_1]} \quad (1)$$

by taking both the magnetization density and anisotropy into consideration. The simulated results are shown in Fig. 4 (the hollow circles and dashed curve). One can see that they agree fair well with the measured results.

5. Discussion

By precisely controlling the film thickness, the influence of thickness on Curie temperature can be excluded in our study. In addition to the coverage, the Curie temperature can be altered by the magnetization density as well as the magnetic anisotropy of the deposited films. The origins that affect remanence and coercivity for ultrathin films at different deposition temperatures may be the following: crystalline structure such as lattice distortion and strain relaxation, chemical structure due to interdiffusion between the magnetic overlayer and non-magnetic substrate, and morphology of the systems. To clarify these possible contributions, further analyses are indispensable.

By performing the LEED $I(E)$ measurements, the average vertical interlayer distance $a_\perp$ can be determined in the kinematic approximation [17,18]. Fig. 5 shows the vertical interlayer distance of 2 ML Co films for a series of deposition temperatures. Due to the lattice mismatch between Co overlayers and Cu substrate at the interface, there exists the lattice distortion along the [1 0 0] direction perpendicular to the film surface. For this reason, $a_\perp$ of Co in Co/Cu(1 0 0) is slightly less than that of Cu single crystal, as indicated in Fig. 5. No significant deviation on $a_\perp$ can be observed for all the deposition temperatures within the accuracy of ± 0.02 Å. It indicates that there is no strain variation for the films at all deposition temperatures. The total volume of the Co atoms also keep unchanged for all the films at different growth temperatures. Furthermore, the variation of the AES ratio of Co to Cu at different deposited-temperature films is insignificant. It means that Co atoms have not diffused deeply into the Cu substrate. This agrees with the previous studies that the onset temperature of the interdiffusion in Co/Cu(1 0 0) system was found to be >400 K [13,21,22].

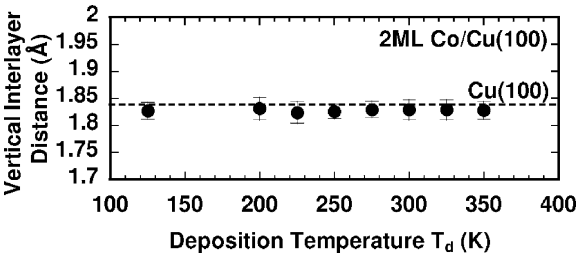

Fig. 5. Vertical interlayer distance ($a_\perp$) of the 2 ML Co films for different deposition temperatures. The dashed line indicates $a_\perp$ for the Cu substrate along [1 0 0] direction. All the measurements were made at 200 K after the films being deposited.

Since neither crystalline nor chemical structure was obviously altered by varying deposition temperature, the variation of the Kerr signal as well as anisotropy and, correspondingly, the Curie temperature should be driven by other origins. As mentioned above, the MEED intensities in Fig. 1 as well as LEED patterns of our experiments suggest that the films deposited at lower temperature reveal pronounced roughness in comparison to those deposited at higher temperature. As a result, the Curie temperature of Co/Cu(1 0 0) films is apparently modified by their morphology. In more detail, the rougher morphology of the films

implies the larger ratio of surface to volume atoms due to the island formation. The total magnetization densities increase since the magnetic moments for the Co atoms at the island edge are enhanced due to the reduced coordinate number in comparison to the bulk atoms, as already indicated in the previous studies [29–33]. Furthermore, Weber et al. [34] reported the morphology-induced oscillations of the magnetic anisotropy in Co/Cu(1 0 0). The authors observed that the variation of the morphology resulting from the alternation between filled and incompletely filled atom layers during the film deposited could also cause the change in magnetic anisotropy of Co/Cu(1 0 0).

6. Conclusion

By conducting the systematic investigation, we studied the correlation between the deposition temperature and Curie temperature for the ultrathin Co/Cu(1 0 0) films. The dramatic enhancement of Curie temperature was observed for the film with variation of deposition temperatures from 340 to 275 K. In addition, the remanent Kerr signal as well as coercive field for the films at lower deposition temperatures were found to be larger than those at higher deposition temperatures. Our further analysis shows that the enhanced magnetization density and anisotropy are responsible for the increase of Curie temperature of the films. It indicates that the Curie temperature of ultrathin films is strongly influenced not only by the thickness of the film, but also by the temperature at which the film was deposited. The surface morphology variation ar different deposition temperatures, which affects the surface magnetic moment as well as anisotropy, plays an important role on this Curie temperature enhancement upon cryogenic deposition.

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References

[1] J. Thomassen, F. May, B. Feldmann, M. Wuttig, H. Ibach, Phys. Rev. Lett. 69 (1992) 3831.
[2] D. Li, M. Freitag, J. Pearson, Z.Q. Qiu, S.D. Bader, Phys. Rev. Lett. 72 (1994) 3112.
[3] S. Müller, P. Bayer, C. Reischl, K. Heinz, B. Feldmann, H. Zillgen, M. Wuttig, Phys. Rev. Lett. 74 (1995) 765.
[4] R.D. Ellerbrock, A. Fuest, A. Schatz, W. Keune, R.A. Brand, Phys. Rev. Lett. 74 (1995) 3053.
[5] R. Allenspach, A. Bischof, Phys. Rev. Lett. 69 (1992) 3385.
[6] S.H. Lu, J. Quinn, D. Tian, F. Jona, P.M. Marcus, Surf. Sci. 209 (1989) 364.
[7] G.L. Krasko, G.B. Olson, J. Appl. Phys. 67 (1990) 4570.
[8] U. Gradmann, Magnetism in ultrathin transition metal films, in: K.H.J. Buschow (Ed.), Handbook of Magnetic Materials, vol. 7, Elsevier Science Publishers, Amsterdam, 1993, pp. 1–96, Ch. 1.
[9] M. Zheng, J. Shen, P. Ohresser, C.V. Mohan, M. Klaua, J. Barthel, J. Kirschner, J. Appl. Phys. 85 (1999) 5060.
[10] M.T. Kief, G.J. Mankey, R.F. Willis, J. Appl. Phys. 69 (1991) 5000.
[11] G.J. Mankey, M.T. Kief, R.F. Willis, J. Vac. Sci. Technol. A 9 (1991) 1595.
[12] H.P. Oepen, S. Knappmann, W. Wulfhekel, J. Magn. Magn. Mater. 148 (1995) 90.
[13] C.M. Schneider, P. Bressler, P. Schuster, J. Kirschner, Phys. Rev. Lett. 64 (1990) 1059.
[14] C. Domb, J. Phys. A 6 (1973) 1296.
[15] F.J. Himpsel, J.E. Ortega, G.J. Mankey, R.F. Willis, Adv. Phys. 47 (1998) 511–597.
[16] M.-T. Lin, J. Shen, W. Kuch, H. Jenniches, M. Klaua, C.M. Schneider, J. Kirschner, Phys. Rev. B 55 (1997) 5886.
[17] M. Zharnikov, A. Dittschar, W. Kuch, C.M. Schneider, J. Kirschner, Phys. Rev. Lett. 76 (1996) 4620.
[18] M.-T. Lin, J. Shen, W. Kuch, H. Jenniches, M. Klaua, C.M. Schneider, J. Kirschner, Surf. Sci. 410 (1998) 290.
[19] J.J. de Miguel, A. Cebollada, J.M. Gallego, S. Ferrer, R. Miranda, C.M. Schneider, P. Bressler, J. Garbe, K. Bethke, J. Kirschner, Surf. Sci. 211/212 (1989) 732.
[20] M.-T. Lin, W.C. Lin, C.C. Kuo, C.L. Chiu, Phys. Rev. B 62 (2000) 14268.
[21] L. Gonzalez, R. Miranda, M. Salmerón, J.A. Vergés, F. Ynduráin, Phys. Rev. B 24 (1981) 3245.
[22] H. Li, B. Tonner, Surf. Sci. 237 (1990) 141.
[23] J.A. Stroscio, D.T. Pierce, R.A. Dragoset, Phys. Rev. Lett. 70 (1993) 3615.
[24] Q.Y. Jin, R. Vollmer, H. Regensburger, J. Kirschner, J. Appl. Phys. 85 (1999) 5288.
[25] S.T. Bramwell, P.C.W. Holdsworth, J. Phys.: Condens. Matter 5 (1993) L53.
[26] S. Chikazumi, S.H. Charap, Physics of Magnetism, John Wiley & Sons, New York, 1964.

[27] R.M. White, Quantum Theory of Magnetism, Springer-Verlag, Berlin Heidelberg, 1983.
[28] R.P. Erickson, D.L. Mills, Phys. Rev. B 43 (1991) 11527.
[29] P. Srivastava, F. Wilhelm, A. Ney, M. Farle, H. Wende, N. Haack, G. Ceballos, K. Baberschke, Phys. Rev. B 58 (1998) 5701.
[30] W. Clemens, T. Kachel, O. Rader, E. Vescovo, S. Blügel, C. Carbone, W. Eberhardt, Solid State Commun. 81 (1992) 739.
[31] M. Tischer, O. Hjortstam, D. Arvanitis, J.H. Dunn, F. May, K. Baberschke, J. Trygg, J.M. Wills, B. Johansson, O. Eriksson, Phys. Rev. Lett. 75 (1995) 1602.
[32] A.B. Shick, D.L. Novikov, A.J. Freeman, J. Appl. Phys. 83 (1998) 7258.
[33] Q.Y. Jin, H. Regensburger, R. Vollmer, J. Kirschner, Phys. Rev. Lett. 80 (1998) 4056.
[34] W. Weber, C.H. Back, A. Bischof, C. Würsch, R. Allenspach, Phys. Rev. Lett. 76 (1996) 1940.