

## Confinement mechanism for strong temperature dependence of the interlayer exchange coupling in Co/Cu(001)

J. d'Albuquerque e Castro,\* J. Mathon, Murielle Villeret, and A. Umerski<sup>†</sup>  
*Department of Mathematics, City University, London EC1V 0HB, United Kingdom*

(Received 26 January 1996)

It is shown, within the quantum-well theory of the exchange coupling  $J$  in magnetic multilayers, that the temperature dependence of  $J$  is governed not only by the spacer Fermi surface but also by the confining strength of the ferromagnetic layers. The confinement mechanism is first explored for an exactly solvable parabolic-band model and cases in which it leads to a very strong temperature dependence of  $J$  are identified. Finally, it is demonstrated, using fully realistic tight-binding bands, that the confinement mechanism dominates  $J(T)$  in Co/Cu/Co(001). [S0163-1829(96)02516-6]

It is generally believed that the temperature dependence of the exchange coupling  $J(T)$  between two magnetic layers separated by a nonmagnetic spacer layer is determined entirely by the properties of the bulk spacer layer Fermi surface (FS). The ferromagnetic layers are assumed to play no role as long as the temperature is low compared with the Curie temperature, which is usually the case under experimental conditions. This belief is based on the results of the quantum well theory with complete confinement in an infinitely deep potential well<sup>1</sup> and Ruderman-Kittel-Kasuya-Yosida (RKKY) theory<sup>2</sup> of the oscillatory exchange coupling. Both of these theories predict that  $J(T)$  is determined solely by the velocity of magnetic carriers at the extremal points of the bulk spacer FS. However, the observed temperature dependence of the exchange coupling in Cu-based trilayers<sup>3,4</sup> is much stronger than predicted by the conventional FS mechanism for Cu.<sup>2</sup>

We show that the existing theories fail to explain the observed  $J(T)$  because there is an additional mechanism of the temperature dependence of the interlayer exchange coupling which has not been considered in any of the previous theoretical treatments. This new mechanism of  $J(T)$ , implicit in the quantum well theory, depends not only on the spacer FS but also on the degree of confinement of magnetic carriers in the spacer quantum well. We shall refer to it as the confinement mechanism of  $J(T)$ . We will show that the confinement mechanism is particularly important in Cu-based multilayers.

The physical origin of the confinement mechanism of  $J(T)$  can be easily understood. The coupling strength in the quantum well theory depends on the magnetic contrast, i.e., the difference between the degrees of confinement of the majority- and minority-spin carriers in the spacer quantum well. That in turn depends on the position of the Fermi level  $E_F$  in the well. If the magnetic contrast varies rapidly with  $E_F$ , then a small temperature-induced shift of the chemical potential  $\mu$  must necessarily lead to a strong dependence of  $J$  on  $T$ . However, it is far from obvious that a very strong dependence of the magnetic contrast on  $E_F$  occurs in real systems. In fact, the magnetic contrast for confinement in an infinitely deep quantum well<sup>1</sup> and also in the opposite limit of an infinitely weak confinement (RKKY) is strictly independent of  $E_F$ , and this is the reason why the confinement mechanism is ineffective in these two most commonly used theoretical models.

The purpose of this contribution is to show that there are magnetic multilayers, including the highly relevant class of Cu-based systems, in which the confinement mechanism is so important that it dominates the total temperature dependence of the coupling. We shall first clarify the origin of the confinement mechanism for a parabolic-band model since it is exactly solvable. A much more interesting case occurs when complete confinement is due to hybridization. This is precisely what happens in Cu-based systems<sup>5</sup> and we will show both analytically and numerically, using fully realistic tight-binding bands, that the confinement mechanism dominates  $J(T)$  in the archetypal Co/Cu(001) system.

Assuming the force theorem, we define (see, e.g. Ref. 5) the exchange coupling between two semi-infinite ferromagnets separated by a nonmagnetic spacer of thickness  $L$  by  $J(L) = \{[\Omega^\uparrow(L) + \Omega^\downarrow(L)]_{\text{FM}} - [\Omega^\uparrow(L) + \Omega^\downarrow(L)]_{\text{AF}}\}/A$ . Here,  $A$  is the surface area and  $\Omega_{\text{FM(AF)}}^\sigma$  is the thermodynamic potential of carriers of spin  $\sigma$  in the ferromagnetic (antiferromagnetic) configuration of the magnetic layers.

Consider a parabolic-band model with complete confinement of minority-spin electrons in a spacer quantum well of depth  $V$  and width  $L$ , and with a perfect match between the ferromagnet and spacer bands in the majority-spin channel. The exchange coupling in such a model is given by<sup>6</sup>  $J(L) = [\Omega^\downarrow(L) - \Omega^\downarrow(\infty)]_{\text{FM}}/A$ , where  $\Omega^\downarrow(L)$  is measured relative to a reference state with constant electron densities appropriate to the bulk ferromagnet and bulk spacer.<sup>6</sup> We showed in Ref. 6 that the coupling for a parabolic-band model can be obtained from the derivative of the thermodynamic potential with respect to the spacer thickness. The derivative  $d\Omega^\downarrow(L)/dL$  is, in turn, expressed in terms of the one-electron Green's function at the edge of the well  $G(L, L, E, \vec{k}_\parallel)$ :

$$\frac{d\Omega^\downarrow(L)}{dL} = (V/\pi) \sum_{\vec{k}_\parallel} \int_{-\infty}^{+\infty} \text{Im}G(L, L, E, \vec{k}_\parallel) f(E) dE, \quad (1)$$

where  $\vec{k}_\parallel$  is the wave vector parallel to the layers and  $f(E)$  is the Fermi function. For electrons confined in a finite well, the spectral density  $-(1/\pi)\text{Im}G(L, L, E, \vec{k}_\parallel)$  is a sequence of  $\delta$  functions,<sup>6</sup>

$$-(1/\pi)\text{Im}G(L, L, E, \vec{k}_{\parallel}) = (k_{\perp}/V) \sum_n \delta(k_{\perp}L + \psi - n\pi), \quad (2)$$

where  $k_{\perp} = [2m(E - E_{\parallel})/\hbar^2]^{1/2}$  is the perpendicular wave vector in the spacer, the energy is measured from the bottom of the well, and the phase  $\psi$  is given by<sup>6</sup>

$$\tan(\psi) = 2[E_{\perp}(V - E_{\perp})]^{1/2}/(V - 2E_{\perp}). \quad (3)$$

Here,  $E_{\perp} = E - E_{\parallel}$  and  $E_{\parallel} = \hbar^2 k_{\parallel}^2/2m$ .

In the context of the temperature dependence of the coupling, the most important feature of confinement in a finite well is a strong energy dependence of the phase  $\psi$  of the spectral density. This occurs near the bottom and top of the spacer quantum well where the derivative  $\partial\psi/\partial E$  tends to infinity. One can prove analytically that qualitatively the same behavior of the phase is obtained also in a single-orbital tight-binding model.<sup>7</sup> We will show that a rapid variation of the phase of the spectral density with energy provides a new mechanism for a strong temperature dependence of the coupling.

Since  $\text{Im}G$  is clearly a periodic function of  $L$  with a period  $\pi/k_{\perp}$ , we can Fourier analyze it in  $L$  and then perform the trivial integral with respect to  $L$ , which yields

$$J(L) = \frac{1}{A\pi} \text{Re} \sum_{p=1}^{\infty} \sum_{\vec{k}_{\parallel}} \int_{-\infty}^{+\infty} i(p^{-1})f(E) \times e^{2ip[k_{\perp}(\vec{k}_{\parallel}, E)L + \psi(\vec{k}_{\parallel}, E)]} dE. \quad (4)$$

As discussed in Ref. 6 we include in Eq. (4) only the oscillatory components  $p=1, 2, \dots$  since  $p=0$  corresponds to the constant reference term  $\Omega^{\downarrow}(\infty)$ .

The temperature dependence of the coupling arises from a cutoff of the energy integral at  $E \approx \mu$ , which is controlled by the Fermi function in Eq. (4). For large spacer thicknesses, all the oscillatory contributions to the energy integral in Eq. (4) cancel except for those coming from the neighborhood of the cutoff at  $\mu$ . Both in the quantum well theory with infinitely strong confinement<sup>1</sup> and in the opposite limit of a very weak confinement (RKKY), one evaluates the energy integral analytically in the complex plane by expanding the perpendicular wave vector  $k_{\perp}(\vec{k}_{\parallel}, E)$  to the first order in energy about  $E = \mu$ .<sup>1,2</sup> This approximation leads to the usual FS mechanism of  $J(T)$  and is asymptotically exact in both these extreme limits since the phase  $\psi$  is independent of the energy (see Refs. 1 and 6).

The crucial point that has previously been overlooked is that one cannot use the same expansion in the intermediate case of complete but relatively weak confinement. In that case, the phase  $\psi$  varies rapidly with the energy when the Fermi level lies close to the top of the confining well and the phase can no longer be approximated by its value at  $E = \mu$  (the case with  $E_F$  at the bottom of the well is of no practical interest). In fact, it has to be treated on the same footing as  $k_{\perp}(\vec{k}_{\parallel}, E)$ , i.e., the whole argument  $k_{\perp}(\vec{k}_{\parallel}, E)L + \psi(\vec{k}_{\parallel}, E)$  of the imaginary exponential in Eq. (4) has to be expanded about the energy cutoff at  $E = \mu$ . The expansion is straightforward and leads to the following result:

$$J(L) = \frac{k_B T}{4\pi^2} \text{Re} \sum_{p=1}^{\infty} \int \int dk_x dk_y \times \frac{e^{2ip[k_{\perp}(\vec{k}_{\parallel}, \mu)L + \psi(\vec{k}_{\parallel}, \mu)]}}{p \sinh\{2\pi p k_B T [L \partial k_{\perp}(\vec{k}_{\parallel}, \mu)/\partial E + \partial\psi(\vec{k}_{\parallel}, \mu)/\partial E]\}}, \quad (5)$$

where we have replaced the sum over  $\vec{k}_{\parallel}$  by an integral that can be evaluated using the stationary phase approximation.<sup>1</sup> For large spacer thickness, the only nonvanishing contributions come from the neighborhood of  $\vec{k}_{\parallel}^0 = 0$  where  $k_{\perp}(\vec{k}_{\parallel}, \mu)$  is stationary. As usual,<sup>1</sup> the perpendicular wave vector  $k_{\perp}(\vec{k}_{\parallel}, \mu)$  is expanded about  $\vec{k}_{\parallel}^0 = 0$  up to second order in  $k_x$  and  $k_y$  and the phase  $\psi$  is approximated by its value at the stationary point. The resulting integrals are Gaussian and can be evaluated analytically. Higher-order corrections in  $\vec{k}_{\parallel}$  have no effect on the temperature dependence of the coupling and can be neglected. The basic stationary phase approximation then yields

$$J = -\frac{k_F k_B T}{4\pi L} \text{Re} \sum_{p=1}^{\infty} \frac{i e^{2ip[k_{\perp}L + \psi]}}{p^2 \sinh[2\pi p k_B T (L \partial k_{\perp}/\partial E + \partial\psi/\partial E)]}. \quad (6)$$

The perpendicular wave vector  $k_{\perp}$ , the phase  $\psi$ , and their derivatives are evaluated at  $\vec{k}_{\parallel} = 0$  and  $E = \mu \approx E_F$ .

It is clear from Eq. (6) that the temperature dependence of the exchange coupling is controlled by two factors in the argument of the hyperbolic sine in Eq. (6). The first factor ( $\partial k_{\perp}/\partial E$ ) depends only on the spacer FS and gives rise to the usual FS mechanism of  $J(T)$  but the second factor ( $\partial\psi/\partial E$ ) is entirely new. It can be seen from Eq. (3) that the new term  $\partial\psi/\partial E$  depends on the degree of confinement of magnetic carriers in the spacer quantum well (position of the Fermi level in the well). Since the degree of confinement is determined by a mismatch between the spacer and ferromagnet bands, it follows that the confinement mechanism of the temperature dependence  $J(T)$  is a joint property of the spacer and ferromagnet. Moreover, the slope of  $\psi$  becomes infinite at the top of the well and, therefore, a very strong temperature dependence  $J(T)$  is expected in the case when the Fermi level lies very close to the top of the confining well.

We have thus demonstrated that, for a parabolic-band model, a very strong temperature dependence of the coupling arises when the magnetic carriers in the spacer quantum well are on the brink of changing from full to partial confinement. It might seem that this situation is rather artificial and it is, therefore, legitimate to ask whether the confinement mechanism can be important in real systems. Rather surprisingly, the answer is yes and we shall now show that it dominates  $J(T)$  in the archetypal Co/Cu(001) system.

The method we have used for calculating the coupling from the derivative of the thermodynamic potential with respect to spacer thickness is not easily generalizable to a discrete lattice and a fully realistic multiorbital tight-binding band structure. We shall, therefore, calculate the temperature dependence of the exchange coupling in Co/Cu(001) using a generalization<sup>8</sup> of the spin current method<sup>9</sup> to a multiorbital

band structure. The exchange coupling between two semi-infinite Co layers separated by  $N$  atomic planes of Cu is expressed,<sup>8,9</sup> in terms of the surface matrix elements of the one-electron Green's function  $G_L^\sigma(j)$  of an overlayer of  $j$  atomic planes of Cu on the left Co slab and the Green's function  $G_R^\sigma(N-j)$  of an overlayer of  $N-j$  atomic planes of Cu on the right Co slab,

$$J = \sum_{\vec{k}_\parallel} \int_{-\infty}^{+\infty} f(E) F(\vec{k}_\parallel, E, N) dE, \quad (7)$$

$$F(\vec{k}_\parallel, E, N) = -\text{ImTr} \frac{1}{\pi} \ln \{ 1 + S^\dagger [G_L^\dagger(j) - G_L^\dagger(j)] \times S^\dagger [G_R^\dagger(N-j) - G_R^\dagger(N-j)] \}.$$

Here,  $S^\dagger = t^\dagger [1 - G_L^\dagger t G_R^\dagger t^\dagger]^{-1}$ ,  $S^\dagger = t [1 - G_R^\dagger t^\dagger G_L^\dagger t]^{-1}$ ,  $t$  is the tight-binding hopping matrix in the spacer, and the trace is over all atomic orbitals. A cleavage plane which separates the trilayer into isolated left and right overlayers is passed between any two neighboring atomic planes  $j$ ,  $j+1$  in the spacer. The overlayer Green's functions in Eq. (7) are determined by the method of adlayers<sup>10</sup> using a tight-binding parametrization with  $s, p, d$  bands and hopping to second nearest neighbors of the first-principle band structures of bulk Cu and ferromagnetic fcc Co.<sup>11</sup> The input in the calculation of the overlayer Green's functions  $G_L^\sigma(j), G_R^\sigma(N-j)$  are the surface Green's functions of the right and left semi-infinite Co slabs, which are generated by the decimation method of Ref. 12

We used two methods to calculate from Eq. (7) the temperature dependence of the coupling: a "brute force" method in which the  $\vec{k}_\parallel$  sum is determined numerically and the energy integral is evaluated in the complex plane by summing over Matsubara frequencies; and an analytic stationary phase method.<sup>1</sup> The first approach yields the exchange coupling at any finite temperature but the numerical effort becomes prohibitive at temperatures much lower than the room temperature  $T_{\text{rm}}$ . The dependence of the coupling on Cu thickness obtained by this method is shown in Fig. 1 for two temperatures  $T = T_{\text{rm}}/4$  (squares) and  $T = 2T_{\text{rm}}$  (circles). Comparing the computed values of the coupling, we find that the temperature dependence of the coupling

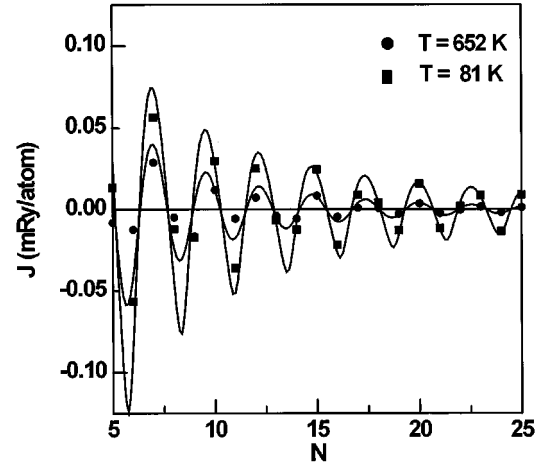


FIG. 1. Dependence of the exchange coupling in Co/Cu(001) on the number of Cu atomic planes determined numerically from Eq. (7) at  $T = 81$  K (squares) and  $T = 652$  K (circles). The continuous curves represent the coupling obtained from the stationary phase formula (8).

$J(T)$  in Co/Cu(001) is much stronger than that expected from the conventional FS mechanism.<sup>2</sup> (It should be noted that our calculated results include both the bilinear and intrinsic biquadratic contributions to the coupling but the intrinsic biquadratic term is negligible.)

In the second approach, we have reconstructed the coupling analytically from Eq. (7) using the stationary phase approximation. The results for the coupling in Co/Cu(001) at  $T = T_{\text{rm}}/4$  and  $T = 2T_{\text{rm}}$  presented in Fig. 1 (continuous curves) are in excellent agreement with those of the brute force method. The analytic approach allows us to determine the reason why the FS mechanism fails to explain the strong dependence of  $J$  on  $T$ . As in the parabolic-band model, the local Green's functions  $G_L^\sigma(j), G_R^\sigma(N-j)$  and, hence the integrand  $F(\vec{k}_\parallel, E, N)$ , are periodic functions of Cu thickness  $N$ . We can, therefore, expand  $F(\vec{k}_\parallel, E, N)$  in a Fourier series in  $N$  and evaluate the  $\vec{k}_\parallel$  sum and the energy integral in Eq. (7) by the same method we had applied to the parabolic-band model. This leads to the following formula for the coupling, which is asymptotically exact for large Cu thickness:

$$J = -\text{Re} \sum_{\vec{k}_\parallel^0} \sum_{p=1}^{\infty} \frac{i |c_p| k_B T \tau m^* e^{2i(pNk_\perp d + \psi_p)}}{2pNd \sinh[2\pi k_B T (pNd \partial k_\perp / \partial E + \partial \psi_p / \partial E)]}. \quad (8)$$

Here,  $|c_p(\vec{k}_\parallel^0, E_F)|$  is the modulus and  $2\psi_p(\vec{k}_\parallel^0, E_F)$  the phase of the Fourier coefficient  $c_p$  of  $F(\vec{k}_\parallel, E, N)$  at a stationary point  $\vec{k}_\parallel^0$  of the Cu FS,  $d$  is the interplanar distance, and  $m^* = |(\partial^2 k_\perp / \partial k_x^2)(\partial^2 k_\perp / \partial k_y^2)|^{-1/2}$ . The perpendicular wave vector  $k_\perp$  and its derivatives are also evaluated at  $\vec{k}_\parallel^0$ . In the relevant [001] direction, the Cu FS has a belly extremum ( $\tau = -i$ ) at  $\vec{k}_\parallel^0 = 0$  and four neck extrema ( $\tau = 1$ ) at  $\vec{k}_\parallel^0 a = (\pm 2.53, \pm 2.53)$ , where  $a$  is the Cu lattice constant. However, it was shown in Ref. 5 that the contribution of the

belly extremum to the coupling is negligible for thick Co layers and it can be thus omitted from our further discussion.

The moduli  $|c_p(\vec{k}_\parallel^0, E_F)|$  and phases  $2\psi_p(\vec{k}_\parallel^0, E_F)$  of the first twenty Fourier coefficients were determined numerically from the computed one-electron Green's functions. As in the parabolic-band model [by expanding the spectral density [Eq. (2)] in a Fourier series in  $L$ ], we find that  $\psi_p \approx p\psi$ , where  $\psi$  is universal. We believe this is due to total confinement. The most striking result, reproduced in Fig. 2, is a very strong energy dependence of the phase  $\psi$  at the neck extre-

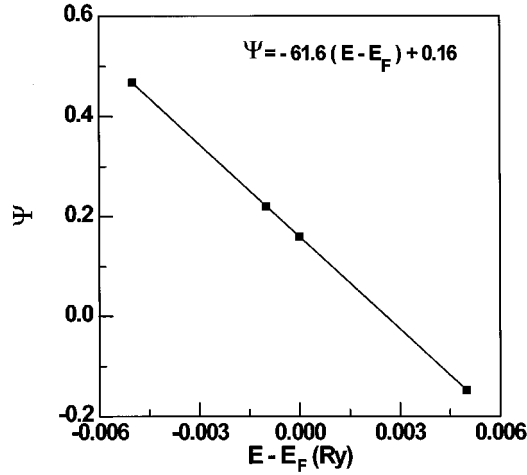


FIG. 2. Energy dependence of the phase of the dominant contribution to the exchange coupling in Co/Cu(001) coming from the Cu Fermi surface necks. The squares are the calculated values and the line is a linear fit through those points.

mum. This is reminiscent of the behavior of the phase in our parabolic-band model with  $E_F$  near the top of the confining well. Since the value  $\partial\psi(\vec{k}_{\parallel}^0, E_F)/\partial E = -61.6 \text{ Ry}^{-1}$  at the neck, deduced from the data shown in Fig. 2, is about a hundred times larger than at the belly extremum, where the confinement of carriers of either spin orientation is only partial, we conclude that it is the confinement of minority electrons in a hybridization gap<sup>5</sup> that causes a strong energy dependence of the phase.

The very large value of  $\partial\psi(\vec{k}_{\parallel}^0, E_F)/\partial E$  is the reason why the FS mechanism fails to explain the temperature dependence of the exchange coupling in Co/Cu(001). The coupling evaluated from the stationary phase formula (8) in which the term  $\partial\psi(\vec{k}_{\parallel}^0, E_F)/\partial E$  is included (continuous curves in Fig. 1) is in an excellent agreement with the computed results. On the other hand, when the confinement mechanism is switched off by setting  $\partial\psi(\vec{k}_{\parallel}^0, E_F)/\partial E = 0$ , we find that  $J(T)$  is far too weak. This is illustrated in Fig. 3 where the temperature dependences  $J(T)$  calculated from Eq. (8) with (curve 1) and without the confinement mechanism (curve 2) are compared for a trilayer with ten atomic planes of Cu. It is clear that the confinement mechanism dominates the temperature dependence  $J(T)$  in Co/Cu(001).

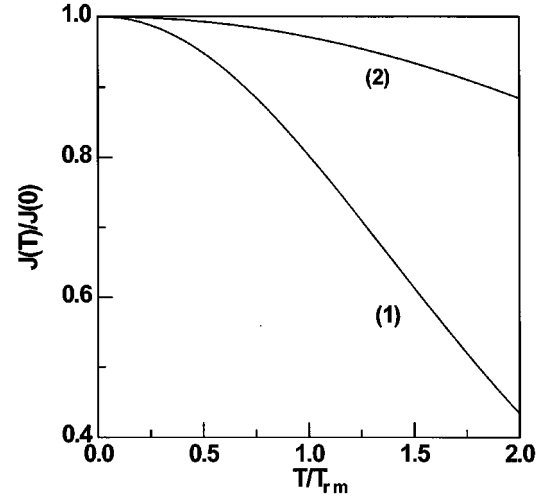


FIG. 3. Comparison of the full temperature dependence of the exchange coupling in Co/Cu(001) calculated from Eq. (8) (curve 1) with the conventional RKKY temperature dependence (curve 2).

The only system for which published experimental results on the temperature dependence of the coupling are available is bcc Fe/Cu(001).<sup>3</sup> Although a comparison with our results for Co/Cu can be merely indicative of the order of magnitude of the effect, it is of interest since we expect the confinement mechanism to be important in Fe/Cu. This is because the minority carriers in bcc Fe/Cu(001) are again fully confined due to hybridization.

Celinski *et al.*<sup>3</sup> measured  $J(T)$  for Fe/Cu with ten atomic planes of Cu between  $T=100 \text{ K}$  and  $T=300 \text{ K}$  and found  $J(100)/J(300) \approx 2$ . Because we assume that the magnetization of the semi-infinite Co layers is independent of  $T$  but Celinski *et al.* estimate that the magnetization of their Fe films is reduced at room temperature by about 15%, it is necessary to scale down their observed factor 2 to  $2(0.85)^2 = 1.44$ . [This assumes (Ref. 5) that the exchange energy is proportional to the square of the Fe film magnetization.] The ratio we obtain from Fig. 3 for Co/Cu is  $J(100)/J(300) = 1.43$ . Such a good agreement is, of course, fortuitous and detailed calculations for bcc Fe/Cu as well as measurements of  $J(T)$  on high-quality Co/Cu(001) samples are required to test the theory quantitatively.

We are grateful to the Engineering and Physical Sciences Research Council, the Royal Society, and CNPq of Brazil for financial support.

\*On leave from Instituto de Física, Universidade Federal Fluminense, 24210-340 Niterói, RJ, Brazil.

†Present address: Department of Mathematics, Imperial College, London SW7 2BZ, U.K.

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