PHYSICAL REVIEW B **72**, 100501(R) (2005)

Peculiar properties of the Josephson junction at the transition from 0 to π state

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(Received 18 April 2005; published 1 September 2005)

It is demonstrated that in the diffusive superconductor-ferromagnet-superconductor (S/F/S) junctions the current-phase relation is practically sinusoidal everywhere except in a narrow region near the $0-\pi$ transition. In this region the second harmonic dominates the scenario of the $0-\pi$ transition. We predict a first-order transition for the S/F/S junctions with a homogeneous F barrier. However, in real junctions a small modulation of the thickness of the barrier may favor the continuous $0-\pi$ transition and the realization of the Josephson junction with an arbitrary ground-state phase difference. The performed calculations of the second-harmonic amplitude provide a natural explanation of the recent contradictory results on the second-harmonic measurements.

DOI: 10.1103/PhysRevB.72.100501 PACS number(s): 74.78.Fk, 74.50.+r

In usual Josephson junctions (JJs) at equilibrium the phase difference of the superconducting order parameter on the two banks is zero.[1] However, the situation may be drastically different for JJs with a ferromagnetic interlayer (S/F/S junctions), where for some intervals of the exchange field h and F-layer thickness d, the ground state corresponds to the phase difference equal to π ("π junctions").[2,3] This phenomenon is related to the damping oscillatory behavior of the Cooper pair wave function in a ferromagnet (for more references and reviews, see Refs. 4 and 5). Experimental evidence of a $0-\pi$ transition in S/F/S ($Nb-Cu_xNi_{1-x}-Nb$) JJs was obtained by Ryazanov *et al.*[6] from the measurements of the temperature dependence of the critical current. The $0-\pi$ transition was signaled by the vanishing of the critical current with the temperature decrease. Such a behavior is observed for a F-layer thickness d close to some critical value d_c. In fact, it simply means that the critical thickness d_c slightly depends on the temperature. The temperature variation serves as a fine tuning and permits to study this transition in detail. Recently, thorough measurements of the critical current were performed in $Nb-Cu_xNi_{1-x}-Nb$ junctions with $x=0.53$ and $d\sim 22$ nm (Ref. 7), and in similar junctions with smaller $x=0.48$ and $d\sim 17$ nm (Ref. 8). The results were contradictory, since the critical current at $0-\pi$ transition in Ref. 7, was zero, while in experiments[8] a small critical current was observed.

In this paper we elaborate a theory describing how the 0 state is transformed into the π state. It is demonstrated that the critical current of the S/F/S JJs does not vanish at the transition and is determined by the second-harmonic term in the current-phase relation. This second-harmonic contribution decreases extremely strongly with the increase of the thickness of the F layer and its exchange field. The corresponding estimate for the critical current at the $0-\pi$ transition in experiments[7] gives the value well below the experimental resolution. On the other hand, for the parameters of S/F/S junctions in Ref. 8, the calculated amplitude of the second harmonic is close to the experimentally measured value. The $0-\pi$ transition is discontinuous for junctions with a homogeneous ferromagnetic barrier. In real S/F/S junctions the modulation of the F-layer thickness may provide a contribution to the second harmonic with the opposite sign.[9,10] If this mechanism prevails then the $0-\pi$ transition would be continuous. This means that by varying the JJ parameters (e.g., temperature) it is possible to obtain the S/F/S junction with an *arbitrary ground-state phase difference*. We also briefly discuss the thermodynamics of the $0-\pi$ transition. Note that previously the current-phase relations in S/F/S junctions were calculated numerically in Ref. 11 and analytically for several special types of the composite SF-FS junctions and short S/F/S junctions in Refs. 5 and 12. However, the theoretical approach to treat analytically the diffusive S/F/S junction with the $0-\pi$ transition (most relevant to the experiment) was lacking.

The current-phase relation for JJs is sinusoidal only near the critical temperature T_c (Ref. 1),

$$j(\varphi) = I_1 \sin \varphi. \tag{1}$$

At low temperature, the higher harmonic terms become more and more important. The calculations of the current-phase relations of the S/F/S junctions in a clean limit indeed reveal strongly nonsinusoidal $j(\varphi)$ dependences at low temperatures.[2,13,14] However, in the experiments[7,8] the ferromagnetic alloys are used as a F layer, and the dirty limit is more appropriate for the description of this case. In such a limit in a normal JJ, if the length d of the weak link exceeds the characteristic length ξ_1 of the decay of the Cooper pair wave function, the critical current is small $j_c \sim \exp(-d/\xi_1)$ and $j(\varphi)$ is practically sinusoidal.[5] We demonstrate that the second-harmonic contribution is very small $\sim\exp(-2d/\xi_1)$. Usually the role of the second harmonic is negligible and hardly observable. However, in S/F/S junctions the first harmonic vanishes at the $0-\pi$ transition and the situation occurs to be very different—the contribution of the second harmonic becomes predominant.

The general current-phase relation

$$j(\varphi) = I_1 \sin \varphi + I_2 \sin 2\varphi \tag{2}$$

corresponds to the following phase-dependent contribution to energy of the JJ:

$$E_J(\varphi) = \frac{\Phi_0}{2\pi c}\left[- I_1 \cos \varphi - \frac{I_2}{2} \cos 2\varphi \right]. \tag{3}$$

If we neglect the second-harmonic term, then the 0 state occurs for $I_1 > 0$. Near a $0-\pi$ transition $I_1 \to 0$ and the second-harmonic term becomes important. The critical current at the transition $j_c = |I_2|$ and if $I_2 > 0$, the minimum energy always occurs at $\varphi=0$ or $\varphi=\pi$ (Fig. 1). In the opposite case ($I_2 < 0$) the transition from 0 to π state is continuous

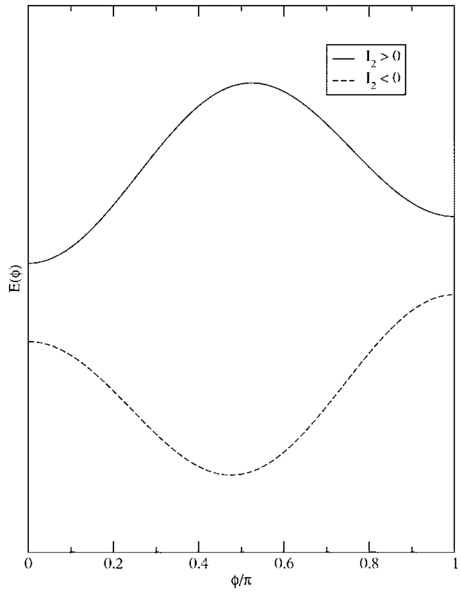

FIG. 1. Schematic plot of the phase-dependent JJ's energy. The case $I_2>0$ corresponds to the discontinuous $0-\pi$ transition while for $I_2<0$ the minimum energy is reached at $0<\varphi<\pi$.

and there is a region where the equilibrium phase difference takes any value $0<\varphi<\pi$. The characteristics of such a "φ junction" are very peculiar.[9]

To describe the properties of the S/F/S junction in the diffusive limit we use the Usadel equations.[15] Recent studies[16,17] revealed a very strong variation of j_c with the F-layer thickness, which implies strong magnetic scattering effects.[18] Assuming the presence of the relatively strong uniaxial magnetic anisotropy in a F layer we may neglect the magnetic scattering in the plane perpendicular to the anisotropy axes (which mixes the spin-up and -down Green's functions) and the Usadel equation for the normal $G(x,\omega,h)$ and anomalous $F(x,\omega,h)$ Green's functions in the F layer is (see, for example, Ref. 4)

$$-\frac{D_f}{2}\left[G(x,\omega,h)\frac{\partial^2}{\partial x^2}F(x,\omega,h)-F(x,\omega,h)\frac{\partial^2}{\partial x^2}G(x,\omega,h)\right]+\left(\omega+ih+\frac{G(x,\omega,h)}{\tau_s}\right)F(x,\omega,h)=0, \quad (4)$$

where the x axis is perpendicular to the junction plane and the F layer corresponds to $-d/2<x<d/2$, D_f is the diffusion constant in the F layer, and τ_s is the magnetic scattering time. In the spatially uniform case, Eq. (4) is equivalent to the one from the Abrikosov-Gorkov theory.[19] Equation (4) must be completed by the boundary conditions at the S/F interface.[20] Below we consider two limiting cases: transparent interfaces and large interface barriers. Moreover, assuming the normal-state conductivity σ_f of the F layer small compared to that of the S layers, $\sigma_s \gg \sigma_f$, we may neglect the influence of the F layer on the S layer, i.e., the Green's functions in the left S layer are $F_s(\omega)=\Delta e^{i\varphi/2}/\Omega$, $G_s(\omega)=\mathrm{sgn}(\omega)\omega/\Omega$, where $\Omega=\sqrt{\omega^2+|\Delta|^2}$ (for the right S layer $\varphi\to-\varphi$).

For transparent interfaces, the boundary conditions[20] express the continuity of the Green's functions. At $T=T_c$ the equation for $F(x,\omega,h)$ is linear and may be easily solved.[4] Just below T_c the nonlinear corrections in (4) are small and we may apply the method similar to that, used in the problem of the nonlinear oscillator.[21] After some calculations we have

$$F(x,\omega)=a\cosh(k_1x)+b\sinh(k_2x)+\frac{b^2-a^2}{32k^2}\left(\frac{1}{D_f\tau_s}+\frac{3}{2}k^2\right)\times[a\cosh(3kx)+b\sinh(3kx)], \quad (5)$$

where the complex wave vectors k_1 and k_2 are determined by the relations

$$k_1^2-k^2=\frac{k^2(5b^2-a^2)}{8}-\frac{(3a^2+b^2)}{4D_f\tau_s},$$

$$k_2^2-k^2=\frac{k^2(5a^2-b^2)}{8}-\frac{(3b^2+a^2)}{4D_f\tau_s}.$$

The nonlinear effects make k_1 and k_2 different from the wave vector $k^2(\omega,h)=2[\omega+ih+\mathrm{sgn}(\omega)/\tau_s]/D_f$ in the solution of the linear equation. The coefficients a and b are determined from the boundary conditions and the current-phase relation may be then directly obtained from the formula for the supercurrent.[15]

In the limit $h\gg T_c$ and in the absence of magnetic scattering the Cooper pair wave function in ferromagnet decays and oscillates at the same characteristic length $\xi_f=\sqrt{h/D_f}$ (Ref. 22). The magnetic scattering leads to the decrease of the decaying length $\xi_1=\xi_f/\sqrt{\alpha+\sqrt{1+\alpha^2}}$ and to an increase of the oscillating length $\xi_2=\xi_f/\sqrt{\sqrt{1+\alpha^2}-\alpha}$, where the dimensionless parameter $\alpha=1/(h\tau_s)$. Note that the product $\xi_1\xi_2=\xi_f^2$ is a constant and independent of α. If the F-layer thickness $d\gtrsim\xi_1$ the expression for the first harmonic reads

$$I_1=j_0\frac{\Delta^2(1+\alpha^2)^{1/4}}{2\sqrt{2}T_c^2}\exp(-d/\xi_1)\left[\sin(d/\xi_2+\Psi)-\frac{\alpha\Delta^2}{96T^2(1+\alpha^2)^{1/2}}\sin(d/\xi_2-\Psi)\right], \quad (6)$$

where $j_0=4\pi T_c eSN(0)D_f/\xi_f$, the angle $\pi/4<\Psi<\pi/2$ is determined by the relation $\tan\Psi=\alpha+\sqrt{1+\alpha^2}$, S is the area of the cross section of the junction, and $N(0)$ is the electron density of state per one spin projection. The amplitude of the first harmonic (6) reveals an oscillatory decay as a function of the F-layer thickness. The sign change of I_1 signals the $0-\pi$ transition. At $T\to T_c$ the critical thickness separating 0 and π phases is $d_c^n(T_c)=(\pi n-\Psi)\xi_2$, which decrease with the decreases with temperature

$$d_c^n(T)=d_c^n(T_c)-\frac{\Delta^2\alpha\xi_2}{96T_c^2(1+\alpha^2)}. \quad (7)$$

The second-harmonic term is much smaller,

$$I_2=-j_0\frac{\Delta^4}{96T_c^4}\exp(-2d/\xi_1)\left[(d/\xi_f)\sin(2d/\xi_2)+\frac{5\sin(2d/\xi_2+\Psi)+3\sin(2d/\xi_2-3\Psi)}{4\sqrt{2}\sin(2\Psi)}\right], \quad (8)$$

and compared with I_1 it contains, in addition to the factor Δ^2/T_c^2, an exponentially small term $\exp(-d/\xi_1)$. The amplitude of $I_2(d)$ also reveals the oscillatorylike dependence similar to $I_1(d)$ but with the decaying and oscillating lengths two times smaller. It may be directly verified that at the $0-\pi$ transition (i.e., when $d=d_c^n$) I_2 is always *positive*.

Now let us consider the limit of large S/F interface barriers which are characterized by the parameter γ_B, related to the S/F boundary resistance per unit area R_b by $\gamma_B=R_b\sigma_f$. In this case the boundary condition reads $\gamma_B(\partial F/\partial x)_{x=d/2}=F_s(x=d/2)G_f^2(x=d/2)$.[20] Performing a similar analysis as in the perfect transparency case ($\gamma_B=0$) we obtain in the limit $d \gtrsim \xi_1$,

$$I_1 = j_0\left(\frac{\xi_f}{2\gamma_B}\right)^2 \frac{\Delta^2}{T_c^2\sqrt{2}(1+\alpha^2)^{1/2}} \exp(-d/\xi_1)\sin(\Psi - d/\xi_2). \quad (9)$$

The second-harmonic term at $\alpha\sim 1$ is of the order $I_2 \sim j_0(\xi_f/\gamma_B)^4 \exp(-2d/\xi_1)$ and also oscillates (with a period $\pi\xi_2$, which is two times smaller than that of I_1), and similar to the transparent interface case is positive at the $0-\pi$ transition.

For small $\alpha \ll 1$, the first $0-\pi$ transition occurs at the F-layer thickness smaller than ξ_f (Ref. 22). In the considered case (assuming $h, \tau_s^{-1} \gg T$) the first $0-\pi$ transition occurs at $d=d_c^0=\xi_f\sqrt{3\alpha}$ and

$$I_1 = j_0\left(\frac{\xi_f}{2\gamma_B}\right)^2 \frac{\Delta^2}{2T_c^2}\frac{d_c^0-d}{\xi_f}. \quad (10)$$

The second-harmonic term at the $0-\pi$ transition is also positive and $I_2 \sim j_0(\xi_f/\gamma_B)^4(\Delta^4/T_c^4\sqrt{\alpha})$. The formulas (9) and (10) are written for $T \lesssim T_c$ but the corresponding analysis is easily generalized for all temperatures. Besides the change of numerical coefficients the expressions for I_1 and I_2 remain the same.

Now we demonstrate how the obtained results permit to understand the controversy in the experimental search of the second harmonic.[7,8] From the thickness dependence of the critical current in the series of Nb$-$Cu$_{0.47}$Ni$_{0.53}$$-$Nb junctions[17,18] we may estimate $\xi_1 \approx 1.4$ nm and $\xi_2 \approx 4.1$ nm. This gives $\xi_f \approx 2.4$ nm, the magnetic scattering parameter $\alpha \approx 1.3$ and the exchange field $h \approx 600$ K. In the experiments[7] the current-phase relation was measured near the second $0-\pi$ transition at $d\approx 22$ nm [the first transition occurs at $d \approx 11$ nm (Ref. 18)]. Therefore we may roughly estimate $I_2/I_1 \sim 0.1\exp(-d/\xi_1) \sim 10^{-8}$, which gives a very small value for $I_2 \lesssim 10^{-11}$ A, well below the experimental threshold. On the other hand, the corresponding estimate for the first $0-\pi$ transition at $d\approx 11$ nm is much more favorable for I_2 observation: $I_2/I_1 \sim 10^{-4}$, and $I_2 \sim 10^{-6}$ A. Therefore, it would be interesting to perform similar measurements on the junctions revealing temperature mediated first $0-\pi$ transition.

In similar junctions, but with smaller Ni concentration $x=0.48$, the second harmonic at the $0-\pi$ transition was reported for the F-layer thickness $d\approx 17$ nm at 1.1 K.[8] This is the first (as a function of d) $0-\pi$ transition. For junctions with $x=0.48$ (Ref. 16) we may roughly estimate $\xi_1\approx 4$ nm, $\xi_2 \approx 9$ nm, the magnetic scattering parameter $\alpha\approx 0.9$, $\xi_f \approx 6$ nm, and $h\approx 100$ K. Extrapolating the expressions (6) and (8) for low temperature we have for the ratio $I_2/I_1 \sim 0.1\exp(-d/\xi_1) \sim 10^{-3}$, which is close to the observed value 3×10^{-3}.[8] On the other hand, for the F-layer thickness $d\approx 19$ nm the $0-\pi$ transition occurs at the temperature 5.3 K and the second harmonic was too small to be observed. Smaller Δ/T ratio and larger F-layer thickness makes this case less favorable for the second-harmonic observation.

We have demonstrated the presence of a small intrinsic second harmonic at the $0-\pi$ transition in S/F/S junctions with a uniform barrier. However, there is another mechanism of the negative second-harmonic generation due to the inhomogeneity of the F-layer thickness.[9] Indeed the roughness of the F layer in the real S/F/S junctions[7,16,17] is of the order of 1 nm. This means that if the characteristic length δl of the thickness variation (along the contact surface) is larger than d, the critical current will vary locally too. On the other hand, if δl is much smaller than the Josephson length λ_J, which for the current density 10^6 A/m^2 (Ref. 17) is of the order of the junction dimension (50×50 μm^2 in Ref. 17), the measured characteristics of the junction will be effectively averaged. At the $0-\pi$ transition we deal with a system where the local current density is alternating $\pm I_1$ and $\overline{I_1}=0$. The resulting local phase variation leads to the appearance of the *negative* second harmonic in the averaged current-phase relation (Refs. 10 and 9) $I_2 \sim -|I_1|(\delta l/\lambda_J)^2$, where λ_J is the Josephson length corresponding to the current density I_1. The 1-nm roughness of the F layer in the experiments[8,16] permits to estimate for $d\approx 17$ nm the value $|I_1| \sim 5\times 10^6$ A/m^2 and $\lambda_J \sim (10-100)$ μm. At the present time there is no information about the characteristic length δl of thickness variation in the studied S/F/S junctions. Taking it as 1 μm for the 10×10 μm^2 junction[16,8] we have $I_2 \sim -5(10^2-10^4)$ A/m^2 while the experimentally observed value is $\sim 3\times 10^4$ A/m^2 and the sign of j_2 is unknown.

Let us now briefly discuss the thermodynamics of the $0-\pi$ transition. Near the transition temperature T_π, the amplitude of the first harmonic I_1 may be considered as a linear function of T, i.e., $I_1=\mu(T-T_\pi)$, while the second-harmonic term as a temperature independent. The phase-dependent contribution to the free energy of junction (3) being $E_J(\varphi) = \Phi_0/2\pi c[-\mu(T-T_\pi)\cos\varphi - (I_2/2)\cos 2\varphi]$. For $I_2>0$ the transition occurs to be I order and at $T>T_\pi$ (0 phase) $\delta F_0 = -(\Phi_0/2\pi c)\mu(T-T_\pi)$, while at $T<T_\pi$, in the π phase $\delta F_\pi = -(\Phi_0/2\pi c)\mu(T_\pi - T)$. Therefore the latent heat of the transition is $q=(\Phi_0/\pi c)\mu T_\pi$. Taking the parameters of the S/F/S junctions,[8,16] we may estimate $\mu T_\pi \sim 10^{-4}$ A and then $q \sim 3\times 10^{-20}$ J. The S/F/S junctions studied in Ref. 18 with the $0-\pi$ transition at $d\approx 11$ nm reveal the parameter μT_π and consequently latent heat, which must be two orders of magnitude larger.

In the case $I_2<0$ the $0-\pi$ transition is continuous and in the interval $-|I_2|<I_1<|I_2|$ the equilibrium phase difference is determined by $\cos\varphi=\mu(T-T_\pi)/|I_2|$. The specific heat of this $\varphi-$ phase is $\delta C=(\Phi_0/2\pi c)T(\mu^2/2|I_2|)$. Therefore we may expect on experiment an increase of the specific heat by δC in the narrow temperature region $T_\pi - \mu|I_2| < T < T_\pi + \mu|I_2|$. If we suppose that the $0-\pi$ transition observed in Ref. 8 is continuous, we may estimate δC in the φ phase. Taking the experimental value of $I_2\sim 3$ μA we have $\delta C \sim 10^{-18}$ J/K=aJ/K. The recent precise measurements of the specific heat of the superconducting microrings[23] demonstrated the possibility to register a specific-heat variation of the order of 0.1 aJ/K per ring.

It may be easily verified that (independently on the sign of I_2 and then on the scenario of the transition) the critical current varies near the transition temperature ($|I_1| \ll |I_2|$) as $I_c(T)=|I_2|(1+\mu|T-T_\pi|/\sqrt{2}|I_2|)$. The temperature dependence is linear and the slope dI_c/dT near T_π is $1/\sqrt{2}$ times smaller than that far way from T_π.

At $T=T_\pi$ the current-phase relation is $j(\varphi)=I_2 \sin 2\varphi$ and then the ac Josephson effect would imply the frequencies two times larger $\omega \to 2\omega = 2[(2e/\hbar)V]$. Note that this circumstance was responsible for the observation of the half-integer Shapiro steps at $0-\pi$ transition in experiments[8] and provided an additional proof of the nonzero critical current at $T=T_\pi$. The superconducting quantum interference device (SQUID) with such junctions would have the periodicity $\Phi_0/2$ on the magnetic flux.

The structure of the soliton (Josephson vortex) is rather peculiar for the long (along the y-axis) junction near a $0-\pi$ transition. For $I_2>0$, the phase distribution is determined by the following equation:

$$\frac{d^2\varphi}{dy^2} = \frac{1}{\lambda_{J0}^2}(\varepsilon \sin\varphi + \sin 2\varphi), \tag{11}$$

where $\lambda_{J0}^{-2}=c\Phi_0 S/(8\pi^2 t|I_2|)$, $\varepsilon=I_1/I_2$, and t is the effective junction thickness. The soliton-type solution is

$$\varphi = \arccos\left(1 - \frac{2(1+\varepsilon)}{1+\varepsilon\cosh^2(\sqrt{1+\varepsilon}\,y/\lambda_{J0})}\right). \tag{12}$$

The variation of the shape of the soliton is presented in Fig. 2. Approaching the transition the central part of the soliton with $\varphi \approx \pi$ grows and finally at $I_1=0$ the system has two degenerate ground states $\varphi=0$ and $\varphi=\pi$. If $I_2<0$, the φ junction is realized in the interval $-|I_2|<I_1<|I_2|$. The S/F junctions near the temperature $T=T_\pi$ could provide an excellent possibility to study the unusual properties[9,10] of these junctions.

In summary, we present an analytical solution of the problem of the second-harmonic contribution to the current-phase relation of the S/F/S JJs in a diffusive limit in the presence of uniaxial magnetic scattering. Note that very recently the case of the isotropic magnetic scattering has been studied numerically in Ref. 24 with qualitatively similar results. An important conclusion of our work is that the $0-\pi$ transition is discontinuous for the S/F/S JJs with a homogeneous F-barrier but may be continuous in real junctions with modulated F-layer thickness. In the latter case a very special φ junction exists in the transition region. The modern microcalorimetric technique could be used for the experimental study of the thermodynamics of the $0-\pi$ transition and determines its type.

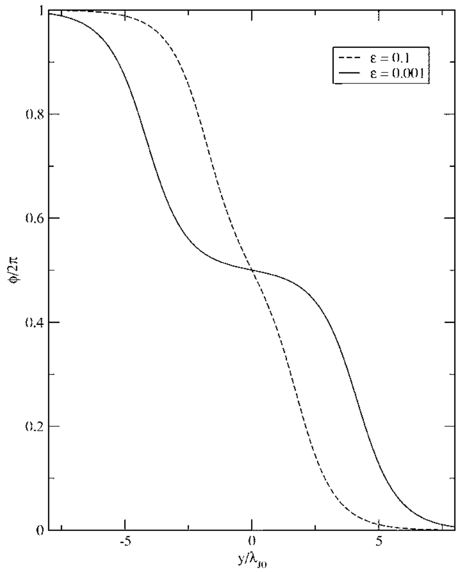

FIG. 2. The change of the form of the Josephson vortex at the discontinuous $0-\pi$ transition. The parameter $\varepsilon=I_1/I_2$ vanishes at the transition.

The author thanks M. Houzet for valuable comments and helpful suggestions. Useful discussions with M. Kupriyanov, O. Bourgeois, M. Faure, E. Goldobin, M. Kulic, Ch. Meyers, V. Ryazanov, and D. Van Harlingen are also gratefully acknowledged.

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