

## de Haas–van Alphen study in the superconducting state of $\text{YNi}_2\text{B}_2\text{C}$

G. Goll\*

*Grenoble High Magnetic Field Laboratory, Max-Planck-Institut für Festkörperforschung  
and Centre National de la Recherche Scientifique, B.P. 166, F-38042 Grenoble Cedex 9, France*

M. Heinecke

*1. Physikalisches Institut, Universität Göttingen, D-37073 Göttingen, Germany*

A. G. M. Jansen, W. Joss, L. Nguyen, and E. Steep

*Grenoble High Magnetic Field Laboratory, Max-Planck-Institut für Festkörperforschung  
and Centre National de la Recherche Scientifique, B.P. 166, F-38042 Grenoble Cedex 9, France*

K. Winzer

*1. Physikalisches Institut, Universität Göttingen, D-37073 Göttingen, Germany*

P. Wyder

*Grenoble High Magnetic Field Laboratory, Max-Planck-Institut für Festkörperforschung  
and Centre National de la Recherche Scientifique, B.P. 166, F-38042 Grenoble Cedex 9, France*

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In the borocarbide superconductor  $\text{YNi}_2\text{B}_2\text{C}$  the de Haas–van Alphen (dHvA) oscillations have been observed continuously at the upper critical field  $B_{c2}$  for the transition from the normal into the superconducting state. In the normal state above  $B_{c2} = 10.5$  T six different frequencies were found in magnetic fields up to 23 T and for  $\Theta = 14.4^\circ$ . The smallest orbit with  $F_\alpha = 511$  T is present down to about 4 T, but with an additional attenuation of the dHvA signal in the superconducting state relative to an extrapolation of the standard Lifshitz-Kosevich formula. The observation of magnetoquantum oscillations over such a wide field range and very close to  $B_{c2}$  enables a detailed discussion of the damping in the context of recent theories of these oscillations in the superconducting state.

The family of borocarbide superconductors  $R\text{Ni}_2\text{B}_2\text{C}$  ( $R$  = rare-earth element) with relatively high transition temperatures has attracted growing interest.<sup>1–3</sup> Superconductivity is observed not only for the nonmagnetic Lu, Y, and Sc compounds, but also for the magnetic Ho, Er, and Tm compounds, which make these systems ideal candidates for the study of the competing effects of superconductivity and magnetism. The knowledge of the electronic structure close to the Fermi energy is a key for the understanding of the superconductivity mechanism in these systems. Band-structure calculations for  $\text{LuNi}_2\text{B}_2\text{C}$  and  $\text{YNi}_2\text{B}_2\text{C}$  revealed a three-dimensional (3D) band structure despite the layered structure with rather complicated band components near the Fermi level.<sup>4–6</sup> A broad  $s$ - $p$  band has been proposed to play the key role in superconductivity rather than a narrow Ni  $3d$  band.<sup>4</sup> This picture is partly supported by recent near-edge x-ray-absorption fine structure measurements,<sup>7</sup> but more information is highly needed.

The de Haas–van Alphen (dHvA) effect, associated with the quantization of electron orbits in a magnetic field is known to be a powerful tool for directly probing the Fermi surface of metals in the normal state as well as in the superconducting state, although with an additional damping of the oscillations compared to the normal state.<sup>8–10</sup> The mechanism of the dHvA effect in the superconducting state is still unclear and controversially discussed by recent theories as well for 2D (Ref. 11) as for 3D metals<sup>12–14</sup> which will be

considered here. Maki and Wasserman and Springford<sup>12</sup> treated the additional damping as due to the quasiparticle scattering at an averaged random flux lattice. Thereby the perturbation approach is valid only close to  $B_{c2}$ . In contrast, Dukan and Tesanovic<sup>13</sup> explained the additional damping in the superconducting state by the gapless nature of the BCS quasiparticle spectrum in high fields related to the periodic structure of the flux lattice which leads to an additional damping factor  $\sim [\max(T, \Gamma)/\Delta]^2$  where  $\Gamma$  is the damping and  $\Delta$  the energy gap. Another approach was given by Miller and Györfy<sup>14</sup> who identified the mechanism of dHvA oscillations in the superconducting state as essentially the same as in the normal state, but the energy gap itself leads to a new damping factor  $\sim aK_1(a)$  where  $a = 2\pi p\Delta/\hbar\omega_c$  and  $K_1(a)$  is a first-order modified Bessel function. In order to clarify this issue, experiments on clean material deep into the superconducting state are necessary.

The present work reports on dHvA experiments on the nonmagnetic  $\text{YNi}_2\text{B}_2\text{C}$  compound which yields the following new results. (1) In the normal state more than ten dHvA frequencies can be resolved corresponding to six extremal Fermi-surface orbits and several harmonics. (2) The quantum oscillations have been observed continuously at the upper critical field  $B_{c2}$  for the transition from the normal into the superconducting state and persist over a wide field range deep into the superconducting state, at least down to fields  $B \approx 0.4B_{c2}$  and at temperatures  $T \approx 0.024T_c$ . (3) Most im-

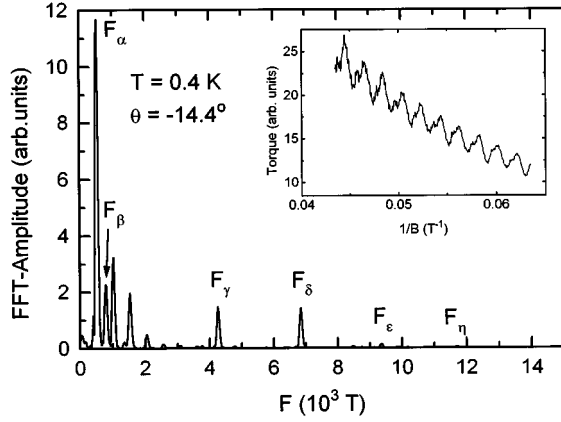


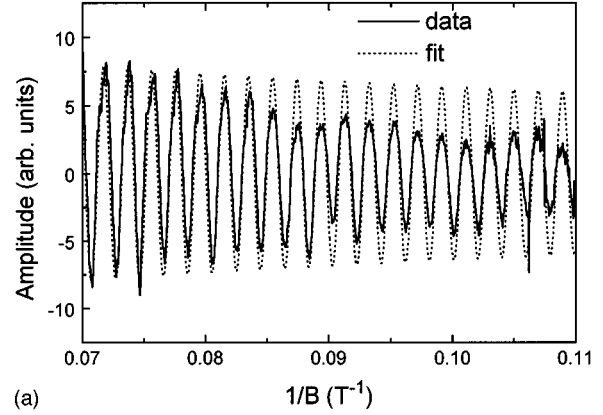
FIG. 1. Fourier spectrum of dHvA oscillations of  $\text{YNi}_2\text{B}_2\text{C}$  in the field range  $15.7 \leq B \leq 23$  T at  $T = 0.4$  K. The inset shows the torque signal vs inverse field  $1/B$  in this field range. The sample was tilted  $\Theta = 14.4^\circ$  off the  $c$  direction in the (110) plane.

portantly, our results provide an unambiguous experimental test of the recent theories of the dHvA effect in the superconducting state. The damping of the dHvA signal in the superconducting mixed state can be explained by the combined influence of both gapped and gapless regions of the Fermi surface.

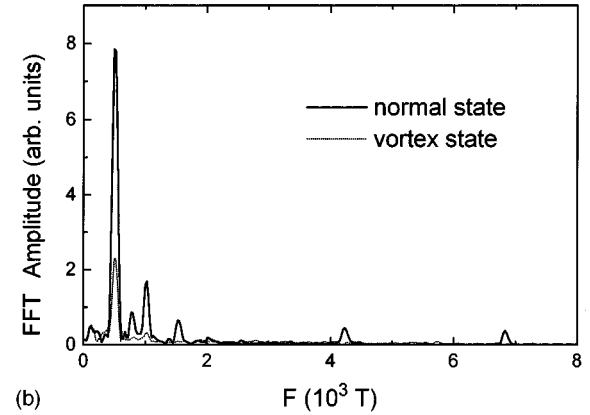
The  $\text{YNi}_2\text{B}_2\text{C}$  sample was a platelet-shaped single crystal with a mass of  $\sim 1$  mg, grown by a high-temperature flux technique with  $\text{Ni}_2\text{B}$  as flux. The superconducting transition temperature  $T_c$  was determined by low-field ac susceptibility which yielded  $T_c = 15.7$  K with a transition width  $\Delta T_c = 0.4$  K (Ref. 15). The residual resistivity ratio of the sample was measured to  $\rho(300 \text{ K})/\rho(17 \text{ K}) = 43$ . The temperature dependence of the upper critical field  $B_{c2}$  was determined in the temperature region  $0.35 < T < 4.2$  K for  $B \parallel c$  which leads to a value  $B_{c2}(0) = 10.6$  T. Therefore, the superconducting coherence length  $\xi_{\text{GL}}$  is  $\xi_{\text{GL}} = \sqrt{\Phi/(2\pi B_{c2})} = 5.5$  nm and the Ginzburg-Landau parameter  $\kappa = 27$  (Ref. 15), proving that  $\text{YNi}_2\text{B}_2\text{C}$  is a type-II superconductor. Measurements of the dHvA effect of this sample in fields up to 12 T by the modulation technique have already been reported.<sup>15</sup>

The torque signal which is proportional to the magnetization multiplied by the applied magnetic field, was measured in a  $^3\text{He}$ -bath cryostat down to 0.4 K using a capacitive cantilever torqueometer fixed on a rotatable sample holder as described in Ref. 16. The sample was glued onto the cantilever by a small amount of grease with  $B \parallel c$  for  $\Theta = 0$  and rotations in the (110) plane. The measurements were performed at the High Magnetic Field Laboratory in Grenoble using a resistive magnet for fields up to 23 T.

We first discuss the normal-state results. Figure 1 shows the spectrum obtained by fast Fourier transformation (FFT) of the data shown in the inset of Fig. 1 measured in the field range  $15.7 \leq B \leq 23$  T and at  $T = 0.4$  K. The sample was rotated by  $\Theta = 14.4^\circ$  off the  $c$  direction in the (110) plane. Eleven frequencies can be resolved for this angle corresponding to six extremal orbits of the Fermi surface and five harmonics. For the specific orientation of the sample the frequencies are  $F_\alpha = 511$  T,  $F_\beta = 784$  T,  $F_\gamma = 4221$  T,  $F_\delta = 6833$  T,  $F_\epsilon = 9385$  T, and  $F_\eta = 11\,700$  T. Moreover, up



(a)



(b)

FIG. 2. (a) dHvA oscillations vs inverse field  $1/B$  in the normal and superconducting state. The fit (dotted line) is an extension of the normal state using the Lifshitz-Kosevich formula with parameters  $F = 510.7$  T,  $m^* = 0.34$ ,  $T_D = 0.264$  K, and  $T = 0.4$  K. (b) Fourier spectrum in the normal state (solid line, field range  $12.7 \leq B \leq 17$  T) and superconducting state (dotted line, field range  $8.3 \leq B \leq 10$  T). The field direction is the same as in Fig. 1.

to the sixth harmonic of the fundamental frequency  $F_\alpha$  can unambiguously be resolved. Preliminary results on the angular dependence of the different frequencies give for  $\Theta = 1^\circ$ , i. e., close to  $B \parallel c$ ,  $F_\alpha = 495 \pm 4$  T,  $F_\beta = 761 \pm 35$  T,  $F_\gamma = 5830 \pm 23$  T,  $F_\delta = 6966 \pm 14$  T, and  $F_\eta = 11210 \pm 40$  T.  $F_\epsilon$  cannot be resolved for  $\Theta < 10^\circ$ . Using the Onsager relation  $F = (\hbar/2\pi e) \cdot A$  the cross-section area  $A$  of the extremal orbits can be calculated. The observed frequencies correspond to cross sections which are 1.5, 2.3, 17.7, 21.1, and 34.0 %, respectively, of the cross-section area  $(2\pi/a)^2 = 315 \text{ nm}^{-2}$  of the Brillouin zone.

We now turn to the main topic of this article, the behavior of the dHvA oscillations in the superconducting state. On passing in the vortex state by lowering the field a distortion of the dHvA signal near  $B_{c2}$  that has been reported for other superconductors exhibiting oscillations in the superconducting state like  $\text{NbSe}_2$  (Ref. 8),  $\text{V}_3\text{Si}$  (Ref. 9), and  $\text{Nb}_3\text{Sn}$  (Ref. 10) is *not* observed. The oscillations persist over a wide field range deep into the superconducting state, at least down to  $B = 4$  T. Figure 2(a) shows the oscillatory part of the torque signal in the field range  $9.1 \leq B \leq 14.3$  T at  $T = 0.4$  K above and below the critical field  $B_{c2} = 10.5$  T for the above-mentioned field direction. The background which was subtracted in Fig. 2(a) is proportional to the static magnetization

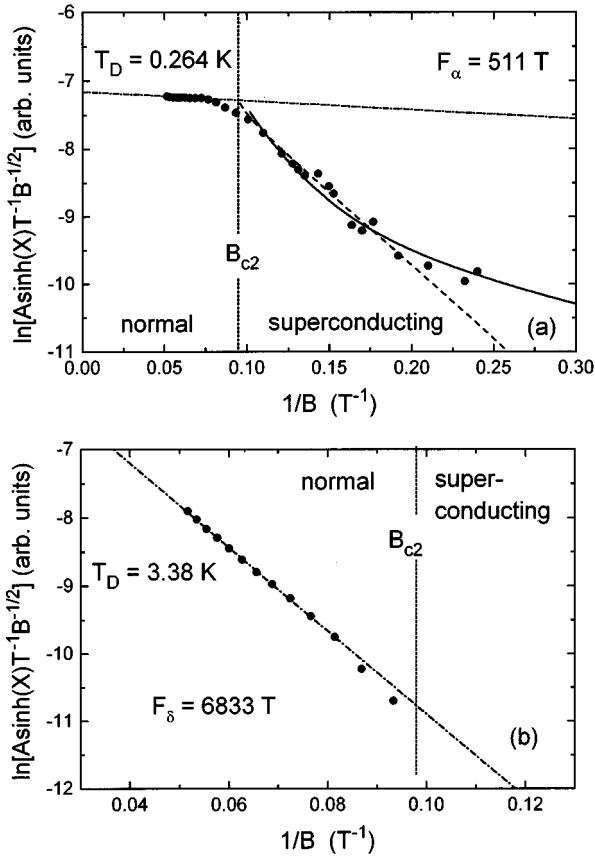


FIG. 3. “Dingle-plot”:  $\ln[A \sinh(X) T^{-1} B^{-1/2}]$  as a function of  $1/B$ , where  $X = 14.69 m^* T/B$  T/K, for the orbits with  $F_\alpha = 511$  T (a) and  $F_\delta = 6833$  T (b). From the slope of the straight (dashed-dotted) line fitted to the data above 13 T a Dingle temperature  $T_D = 0.264$  K (a) and  $T_D = 3.38$  K (b), respectively, was obtained. For the fit curves in the superconducting-state theories by Dukan and Tesanovic (Ref. 13, solid line), and Miller and Györfy (Ref. 14, dashed line) were used. For explanation and parameters see text.

$M(B)$  and shows the usual irreversibility of the magnetization curves of a superconductor below the irreversibility field due to flux pinning, actually proving that the sample is in the superconducting state. Below  $B_{c2}$  an additional damping of the oscillation amplitude is observed relative to an extrapolation of the standard Lifshitz-Kosevich formula<sup>17</sup> which is shown as the dotted curve in Fig. 2(a). The parameters are  $F = 510.7$  T,  $m^* = m_c/m_e = 0.34$ ,  $T_D = 0.264$  K, and  $T = 0.4$  K.

The Fourier analysis of the oscillations in field intervals  $\Delta(1/B) = 0.02$  T<sup>-1</sup> above and below  $B_{c2}$ , respectively, is given in Fig. 2(b). In the normal state the signal is dominated by the frequency  $F_\alpha$  and its harmonics, but  $F_\beta$ ,  $F_\gamma$ , and  $F_\delta$  are also present. In the superconducting state only  $F_\alpha$  can be resolved. This frequency is unchanged between the normal and vortex state which can be seen even more sensitive from Fig. 2(a), where no phase shift is observed near  $B_{c2}$ .

For a detailed analysis of the field dependence of the oscillation amplitude in the range between  $4 \leq B \leq 23$  T the Fourier analysis over field intervals  $\Delta(1/B) = 0.02$  T<sup>-1</sup>, i. e., over ten oscillations of the lowest frequency  $F_\alpha$ , was made. Figure 3(a) [3(b)] shows the FFT amplitude of the oscillations with frequency  $F_\alpha$  ( $F_\delta$ ) in the so-called “Dingle plot,”

i. e.,  $\ln[A \sinh(X) T^{-1} B^{-1/2}]$  as a function of  $1/B$ , where  $X = \alpha m^* T/B$ ,  $\alpha = 2\pi^2 k_B m_e / e \hbar = 14.69$  T/K, and  $m_\alpha^* = m_{c\alpha}/m_e = 0.34$  ( $m_\delta^* = 1.24$ ), as determined by Ref. 15. From the slope of the straight (dashed-dotted) line fitted to the data in the normal state well above  $B_{c2}$  a Dingle temperature  $T_D = 0.264$  K was obtained for the smaller orbit ( $F_\alpha$ ), corresponding to a scattering rate  $\tau^{-1} = 2\pi k_B T_D / \hbar = 2.2 \times 10^{11}$  s<sup>-1</sup>. For the larger orbit ( $F_\delta$ )  $T_D$  is 3.38 K and  $\tau^{-1} = 2.8 \times 10^{12}$  s<sup>-1</sup>. On entering the superconducting state an additional damping occurs which, in fact, starts slightly above  $B_{c2}$  which might possibly be due to superconducting fluctuations and, of course, due to averaging over ten oscillations. While the oscillations with frequency  $F_\alpha$  can be followed deep into the superconducting state, the torque method is not sensitive enough to detect oscillations with frequency  $F_\delta$  below 10 T. For the latter orbit oscillations have been reported for  $0.085$  T<sup>-1</sup>  $\leq 1/B \leq 0.125$  T<sup>-1</sup> by Ref. 15. However, these authors found no obvious change in the slope of the corresponding Dingle plot in this field range and their Dingle temperature  $T_D = 2.81$  K is close to our value obtained in the normal state at higher fields. Therefore, we believe that this orbit is only weakly influenced by superconductivity.

The persistence of dHvA oscillations below  $B_{c2}$  could, in principle, be due to a small portion of the sample remaining normal. However, the sample shows the full diamagnetic signal after cooling in zero magnetic field below  $T_c$  and measuring the magnetization by a superconducting quantum interference device magnetometer in a field of  $10^{-4}$  T, proving that the sample is fully transformed.

Quantum oscillations in the vortex state of type-II superconductors have been investigated by recent theories. Dukan and Tesanovic<sup>13</sup> assign the quantum oscillations to quasiparticles due to the gapless nature of the BCS quasiparticle spectrum in high fields related to the periodic structure of the flux lattice. In other words, the key aspect of this theory is the presence of nodes in the gap broadened either by temperature or scattering while the gap opens at the rest of the Fermi surface. Therefore the amplitude of the oscillatory part of the magnetization is given by  $A = A^G(B, T) + A^{1-G}(B, T)$ .  $A^G$  describes the contribution of the quasiparticles in the gapless regions of the Fermi surface and is equal to the contribution in the normal state reduced by a factor  $G = 2[C \max(T, \Gamma)/\Delta]^2$ . The second term  $A^{1-G}$  takes into account the quasiparticle excitations in the “gapped” regions of the Fermi surface. For the oscillations in the superconducting state of V<sub>3</sub>Si (Ref. 9) the authors achieve good agreement if the damping  $\Gamma$  is calculated self-consistently:  $\Gamma(B) = \sqrt{\Gamma_0 \Delta(B)}/2$ , where  $\Gamma_0 = \pi k_B T_D$  is the normal-state damping and  $\Delta(B) = \Delta_0 \sqrt{1 - B/B_{c2}}$  the thermodynamic averaged gap. For YNi<sub>2</sub>B<sub>2</sub>C the normal-state damping yields  $\Gamma_0 = 0.072$  meV and the energy gap  $\Delta_0 = 2.5$  meV was calculated from  $2\Delta_0/k_B T_c = 3.5$ , consistent with recent measurements of  $\Delta_0$  by point-contact spectroscopy<sup>18</sup> and measurements of the specific heat.<sup>19</sup> Thus  $\Gamma(B) > k_B T$  is true for almost all  $B$  except very close to  $B_{c2}$  and therefore  $G = 2[C\Gamma(B)/\Delta(B)]^2$ . The solid line in Fig. 3(a) is the best possible fit to the data in the superconducting state using  $B_{c2} = 10.5$  T and  $\Delta_0 = 2.5$  meV, with  $C$  being the only fit parameter. Using  $C = 2.4$  the curve de-

scribes the data over a wide field range. Especially at low fields where the first term  $A^G$  is the leading contribution to the amplitude this theory gives an adequate description of the experiment. We note that for the parameter set of  $\text{YNi}_2\text{B}_2\text{C}$  the second term  $A^{1-G}$  cannot be neglected with respect to the first term  $A^G$ , in contrast to the case for  $\text{V}_3\text{Si}$  where the second term can be neglected.

In contrast, Miller and Györfy<sup>14</sup> identify the oscillations in the superconducting state with oscillations of the superconducting ground-state energy itself. They argue that in both the normal and the superconducting state, a magnetic field splits a continuous spectrum into discrete Landau-like peaks. Therefore the superconducting ground state is formed from Cooper pairs whose members individually occupy discrete Landau levels. Thus dHvA oscillations can even exist when the whole Fermi surface is gapped in the superconducting state and the mechanism of dHvA oscillations in the superconducting state is essentially the same as in the normal state, but the presence of the energy gap leads to a new damping factor  $\sim aK_1(a)$  in the limit where  $\Delta \gg k_B T$ .  $K_1(a)$  is a first-order modified Bessel function and  $a$  is given by  $2\pi p \Delta / \hbar \omega_c$  for the  $p$ th harmonic. In the limit when the order parameter  $\Delta$  exceeds the Landau-level spacing  $\hbar \omega_c$ ,  $aK_1(a)$  can be approximated by  $\sqrt{(\pi/2)a} \exp(-a)$ , i.e., by an exponential damping term which is qualitatively the same result as obtained in Ref. 13 for the second term  $A^{1-G}$ , the contribution of the quasiparticles in the gapped regions of the Fermi surface. For  $\text{YNi}_2\text{B}_2\text{C}$  the approximation  $\Delta > \hbar \omega_c$  holds only below 5.3 T. Therefore, we used the exact damping term  $\sim aK_1(a)$  for the comparison with the data. A fit to the data with  $\Delta_0$  and  $B_{c2}$  as fit parameter is given by the dashed line in Fig. 3(a) yielding  $\Delta_0 = 1.2$  meV and  $B_{c2} = 10.5$  T. Clearly, the agreement is quite good, too. However, the experimentally observed damping corresponds to a

gap  $\Delta_0$  which is only  $\approx 1/2$  of the thermodynamic value. This deficiency was already mentioned by Miller and Györfy<sup>14</sup> for other materials, however, the reasons are still unclear.

While neither of the theories are applicable close to  $B_{c2}$ , it should be mentioned that the experimental damping close to  $B_{c2}$  can also be described very well by introducing a field-dependent scattering rate  $\tau_S^{-1} = 2\sqrt{\pi}\Delta^2\Lambda/\hbar v_F$  as an additional damping term,<sup>12</sup> where  $\Lambda = (2e\hbar B)^{-1/2}$  and  $v_F = \sqrt{2\hbar e F}/m_c = 4.2 \times 10^5$  m/s is the Fermi velocity, calculated from the dHvA experiment. The fit (not shown in Fig. 3), which is undistinguishable from the data in the vicinity of  $B_{c2}$ , yields  $B_{c2} = 12.4$  T and  $\Delta_0 = 4.75$  meV.  $\Delta_0$  exceeds the BCS value calculated from  $2\Delta/k_B T_c = 3.5$  by a factor of 2 which seems to be rather unreasonable.

In summary, an experimental study of the dHvA effect in the normal and superconducting state of  $\text{YNi}_2\text{B}_2\text{C}$  has been reported. In the normal state at high magnetic fields six extremal Fermi-surface orbits were found yielding important information on the Fermi surface of the borocarbide system. The quantum oscillations persist deep into the superconducting state with an additional damping much larger than the normal-state damping. The observation of the additional damping over such a wide field range enables a detailed analysis of the damping in context of different recent theories. The analysis favors an explanation that the oscillations in the superconducting state might originate from both gapless regions of the Fermi surface and regions where a gap exists.

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\*Permanent address: Physikalisches Institut, Universität Karlsruhe, D-76128 Karlsruhe, Germany.

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