

# Lawrence Berkeley National Laboratory

## LBL Publications

### **Title**

Electron-Phonon Scattering in Si Doped GaN

### **Permalink**

<https://escholarship.org/uc/item/3zr903bd>

### **Author**

Wetzel, C.

### **Publication Date**

1997

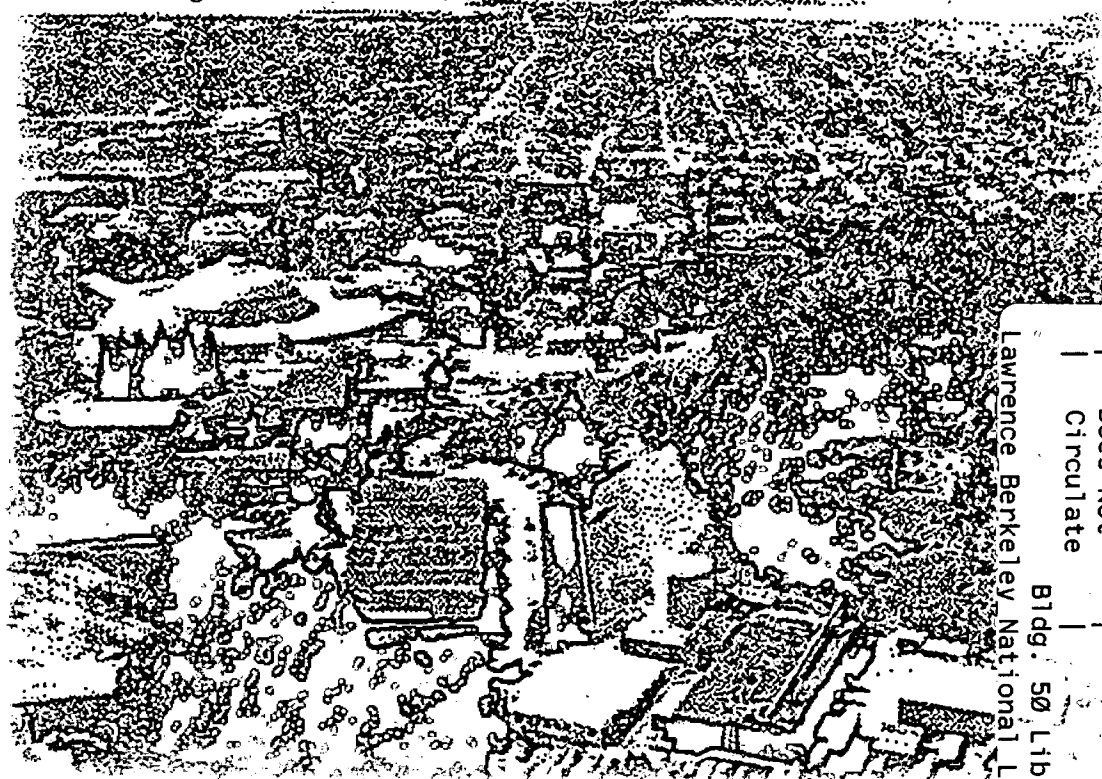


# ERNEST ORLANDO LAWRENCE BERKELEY NATIONAL LABORATORY

## Electron-Phonon Scattering in Si Doped GaN

C. Wetzel, W. Walukiewicz, and J.W. Ager III  
Materials Sciences Division  
Center for Advanced Materials

December 1996  
Presented at the  
*MRS 1996 Fall Meeting*,  
Boston, MA,  
December 2-6, 1996,  
and to be published in  
the Proceedings



REFERENCE COPY |  
Does Not |  
Circulate |  
Bldg. 50 Library - Ref.  
Lawrence Berkeley National Laboratory

## **DISCLAIMER**

This document was prepared as an account of work sponsored by the United States Government. While this document is believed to contain correct information, neither the United States Government nor any agency thereof, nor the Regents of the University of California, nor any of their employees, makes any warranty, express or implied, or assumes any legal responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by its trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof, or the Regents of the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof or the Regents of the University of California.

## **Electron-Phonon Scattering in Si Doped GaN**

C. Wetzel, W. Walukiewicz and J.W. Ager III

**MATERIALS SCIENCES DIVISION**  
Ernest Orlando Lawrence Berkeley National Laboratory  
University of California  
Berkeley, California 94720

December 1996

## ELECTRON - PHONON SCATTERING IN Si DOPED GaN

C. WETZEL, W. WALUKIEWICZ, and J.W. AGER III

Lawrence Berkeley National Laboratory, Materials Science Division, MS 2-200,  
1 Cyclotron Road, Berkeley, CA 94720, USA, C\_Wetzel@LBL.GOV

### ABSTRACT

Phonon-plasmon scattering in non-resonant Raman spectroscopy is used to determine the free electron concentration in Si doped GaN films. For various doping concentration and variable temperature the correlation with magneto-transport data is established. The freeze-out of the carrier concentration at low temperature is thus observed in a purely optical detection scheme. We observe a very long transient time of several hours for the carrier concentration as a reaction to temperature variation. This indicates an indirect capture and emission process with a very small cross section. The value of the Faust-Henry coefficient is determined.

### INTRODUCTION

Assessment of electronic transport properties in group-III nitrides are a basic necessity for both revealing the fundamental physics and as a standard characterization of process parameters. Magneto-transport measurements are typically performed to derive free electron concentration and electron mobilities in Van-de-Pauw or Hall bar electrical contact schemes [1-3]. A convenient purely optical method is to study the interaction of the free electron plasma with the phonons of the lattice in Raman spectroscopy [4-6]. Within a wide range of device relevant doping levels, i.e. free electron concentrations this technique provides a fast contactless method to both determine the parameters on an area defined only by the focal size of the used laser beam or imaging of wafer size areas. Due to the spectroscopic character of the technique further information of the system under investigation is obtained simultaneously.

Established in other binary compound semiconductors, i.e. GaAs the method has been applied to GaN to derive the coupling parameters of the system [5] and determine the carrier concentration in samples under large hydrostatic pressure in diamond anvil cells [6]. In this paper we provide additional information on the sign of the Faust-Henry coefficient and compare data obtained in Raman spectroscopy with magneto-transport results. We investigate a series of Si doped samples at various doping levels and also present the temperature dependence.

### EXPERIMENTAL

A series of nominally undoped and Si doped GaN films were grown by metalorganic vapor phase epitaxy (MOVPE) on (0001) sapphire in two different reactors [3]. Doping and magneto-transport data of the samples are presented in Table I. Raman spectroscopy was performed using 120 mW of the 476.5 nm line of an Ar ion laser and matching narrow band dielectric band blocking filters. Spectra at variable temperature and various polarization were taken in backscattering geometry.

## RESULTS

The coupling of the lattice phonon and the free electron plasmon is described in an oscillator model of the dielectric function  $\varepsilon(\omega)$ .

$$\frac{\varepsilon(\omega)}{\varepsilon_0} = 1 + \frac{\omega_L^2 - \omega_T^2}{\omega_L^2 - \omega^2 + i\omega\Gamma} - \frac{\omega_P^2}{\omega(\omega - i\gamma)}, \quad (1)$$

where  $\omega$  is the photon frequency,  $\omega_L$  and  $\omega_T$  the longitudinal and transverse optical phonon frequencies, respectively.  $\Gamma$  is the phonon damping coefficient.  $\omega_P^2 = e^2 N / (\varepsilon_0 \varepsilon_r m^*)$  is the square of the plasma frequency connecting to the free electron concentration  $N$ . The carrier mobility is related to the electron scattering frequency  $\gamma$ .

Raman scattering in two distinct configurations  $z(x, \bar{x})\bar{z}$  and  $x(z, \bar{z})\bar{x}$  (Cartesian coordinates) geometry is shown in Fig. 1. The selection rules (Fig. 1) [7] are well obeyed and allow for an interpretation of the relative intensities  $I_L$  and  $I_T$  of the  $A_1(\text{LO})$  and  $A_1(\text{TO})$  modes. The ratio can be related to the Faust-Henry coefficient  $C$  by [8,4]:

$$\frac{I_{\text{LO}}}{I_{\text{TO}}} = \left( \frac{\omega_1 + \omega_L}{\omega_1 + \omega_T} \right)^4 \frac{\omega_T}{\omega_L} \left( 1 + \frac{\omega_T^2 - \omega_L^2}{C\omega_T^2} \right)^2, \quad (2)$$

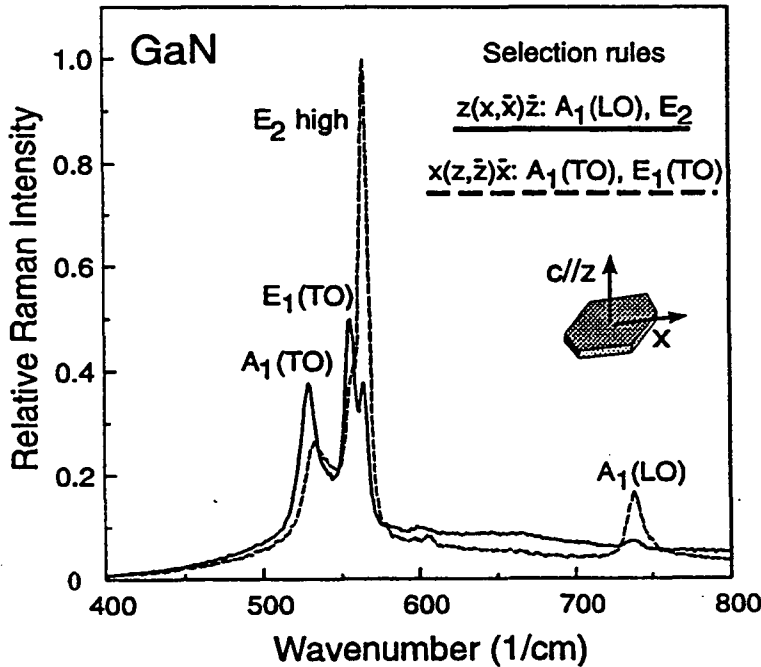


Fig. 1: Non-resonant Raman scattering in backscattering geometry of a low doped GaN/sapphire film in two different orientations along with the selection rules. In this sample the sapphire mode is very weak.

where  $\omega_1$  is the laser frequency. Solving for  $C$  we find two roots  $C \in \{0.4, -5.2\}$ . In a previous work only  $C=0.4$  was found [5].

In order to determine the sign of  $C$  we compare the measured intensities of the  $A_1(\text{LO})$  with the calculated ones [6,5] for samples with various electron concentrations. In Fig. 2 room temperature  $z(x, -)\bar{z}$  spectra scaled to the intensity of the electron concentration independent  $E_2$  mode are presented along with  $N_{\text{Hall}}$  from magneto-transport [3]. We find that for increasing concentration the LO mode decreases in peak height. From a simulation of the lineshape and the absolute intensity compared to the unaffected  $E_2$  mode we

find that this corresponds to the negative value of  $C = -5.2$ .

Values  $N_{Raman}$  from the interpretation of the lineshape are also collected in Table I. The carrier mobility was assumed to be identical to the value from the transport measurement  $\mu_{Hall}$ . We find good agreement for the derived carrier concentrations. The assumption of a constant mobility in both experiments is typically good in the limit of high mobility [5]. For low mobility values typically  $\mu_{Hall}$  shows higher values than optically derived mobilities.

This originates in the different weighing of the scattering events. Similar effects have been described comparing cyclotron resonance, optically detected cyclotron resonance, Shubnikov-de-Haas effect with Hall-effect in GaInAs quantum well systems [9]. In principle for the macroscopic property only the projection of the scattering event along the drift direction matters, whereas for the microscopic detection any scattering direction destroys the particle of the coupled mode. A detailed analysis is currently under way.

In the next step we correlate the phonon-plasmon coupled mode with transport data as a function of temperature. Fig. 3 gives the center of the  $E_2$  mode in the range of 16 K – 95 K. A total shift of  $1.72 \text{ cm}^{-1}$  is found. The linewidth  $3.0 \pm 0.4 \text{ cm}^{-1}$  varies very little. A similar shift of the  $A_1(\text{LO})$  mode and the sapphire mode is found for the little doped sample  $N_{Hall} = 8.6 \times 10^{16} \text{ cm}^{-3}$ . The self energy effects will be considered elsewhere.

A full set of spectra at various temperatures were taken on the higher Si doped sample

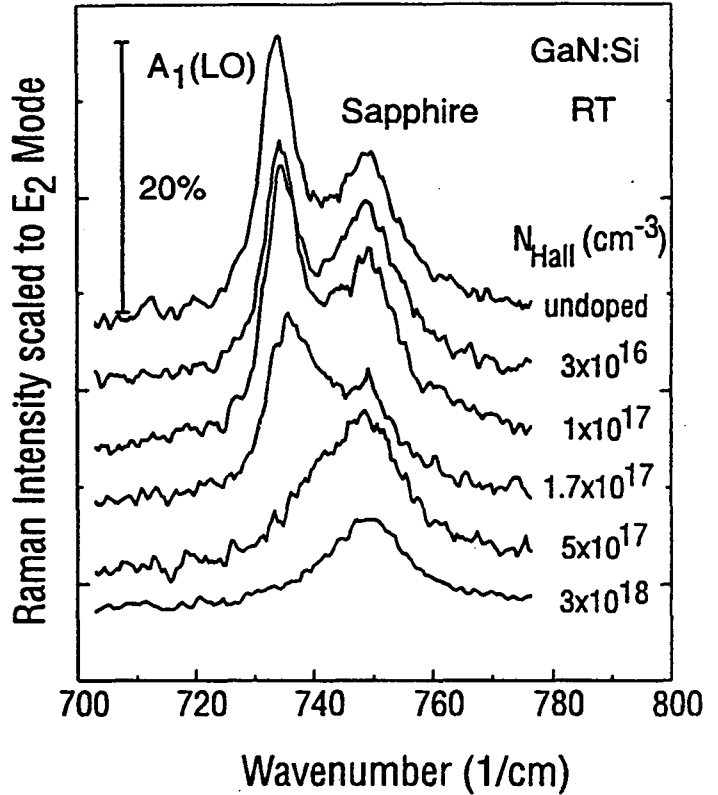


Fig. 2: Raman spectra of GaN films with various Si doping levels scaled to the intensity of the  $E_2$  mode. The lineshape and intensity of the  $A_1(\text{LO})$ -plasmon coupled mode is sensitive to the free electron concentration. From comparison with calculations we find  $C = -5.2$ . In this material an additional sapphire mode at  $750 \text{ cm}^{-1}$  is seen.

Table I: Data for Si doped MOVPE GaN/sapphire films.  $N_{Hall}$  and  $\mu_{Hall}$  refer to free electron concentration and mobility from magneto-transport,  $N_{Raman}$  and  $\mu_{Raman}$  refer to data from the Raman study. All data represent room temperature values.

Sample	A	B	C	D	E	F	G
$N_{Hall} (10^{16} \text{ cm}^{-3})$	'undoped'	3.5	8.6	10	17	50	300
$N_{Raman} (10^{16} \text{ cm}^{-3})$	<5	7	9	4	10	35	--
$\mu_{Hall} (\text{cm}^2/\text{Vs})$	--	--	410	240	190	355	230

$N_{Hall} = 5 \times 10^{17} \text{ cm}^{-3}$  (Fig. 4). The whole temperature cycle consists of a 1.5 h cool down phase, a 17 h settling time and a 3 h warm up phase. The spectra at the various extreme points are displayed in Fig. 5. Starting at room temperature only a weak contribution of the  $A_1(\text{LO})$  mode is found on the background of the sapphire mode. Lowering the temperature a monotonically growing and narrowing LO mode is observed. From a fit to the lineshape of the coupled mode we derive a total decrease in the free electron concentration from  $3.5 \times 10^{17} \text{ cm}^{-3}$  to  $\sim 1 \times 10^{17} \text{ cm}^{-3}$ . During the subsequent settling time without laser irradiation the temperature

varies little however, there is a considerable change observable in the coupled mode. We derive an electron concentration as low as  $\leq 5 \times 10^{16} \text{ cm}^{-3}$ . During the long warm up phase under laser irradiation a similar retardation with a time constant of 2 -- 3 h in the change in the LO mode is observed. Reaching a temperature of 265 K the mode is still very well pronounced superseding the mode right after cool-down.

This indicates that the carrier concentration has not yet reached its equilibrium room temperature concentration. During the whole temperature cycle the correct sample temperature is directly monitored from the position of the  $E_2$  mode.

## DISCUSSION

The temperature induced freeze-out of the free electron concentration is

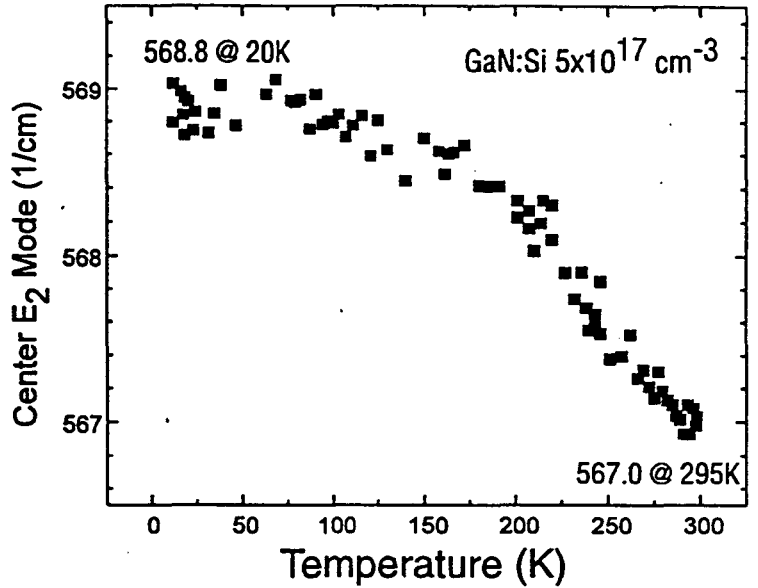


Fig. 3: Center of the  $E_2$  mode as a function of temperature for  $N_{Hall} = 5 \times 10^{17} \text{ cm}^{-3}$ . The absolute position is sample dependent.

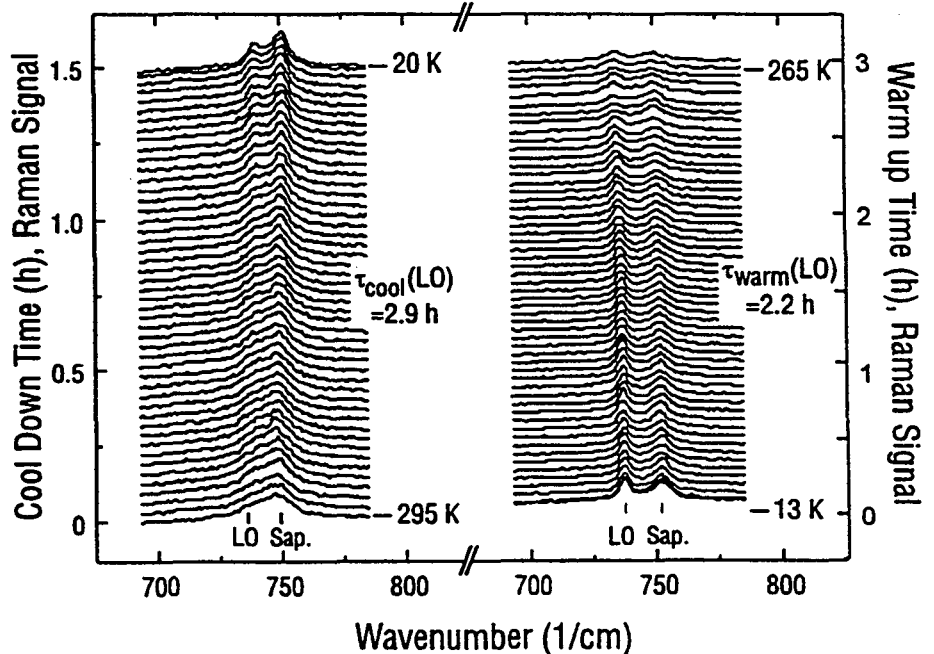


Fig. 4: The  $A_1(\text{LO})$  phonon range at various temperatures. A significant transient time for the carrier concentration, i.e. lineshape is observed. The sample temperature is monitored in parallel by the  $E_2$  mode.



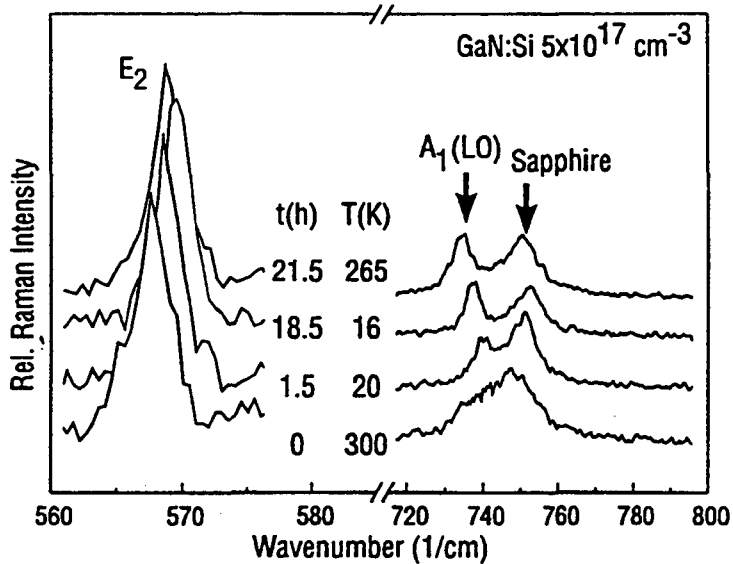


Fig. 5: Raman spectra at the several stages of the temperature cycle ( $N_{Hall}=5 \times 10^{17} \text{ cm}^{-3}$ ). The appearance of the  $A_1(\text{LO})$  mode corresponds to the freeze-out of the carriers.

of the Si donor these long time scales indicate the participation of intermediate steps and possibly reacting partners. Likely candidates are participating acceptors or mobile compensating defects. The typically highly defective crystal structure of epitaxial GaN films offers a high density of possible impurity and complex partners.

These findings provide important hints for a dynamical compensation mechanism active in GaN thin films and possibly also in devices.

## ACKNOWLEDGMENTS

We gratefully acknowledge helpful discussions with Professor E.E. Haller. Part of the work was supported by the Director, Office of Energy Research, Office of Basic Energy Sciences, Materials Science Division of the U.S. Department of Energy under Contract No. DE-AC03-76SF00098.

## REFERENCES

- [1] N. Koide, H. Kato, M. Sassa, S. Yamasaki, K. Manabe, M. Hashimoto, H. Amano, K. Hiramatsu, I. Akasaki, *J. Cryst. Growth*, **115**, 639 (1991).
- [2] P. Hacke, A. Maekawa, N. Koide, K. Hiramatsu, N. Sawaki, *Jpn. J. Appl. Phys.* **1**, **33**, 6443 (1994).
- [3] W. Götz, N.M. Johnson, C. Chen, H. Liu, C. Kuo, W. Imler, *Appl. Phys. Lett.* **68**, 3144 (1996).

directly observed in the lineshape and position of the  $A_1(\text{LO})$  mode. The total variation of the derived concentration is equivalent to the freeze-out behavior observed in the magneto-transport experiment presented before (Sample No 4 in Ref. [3]). This allows for a direct correlation of the two methods to derive free electron concentration in a contactless optical method.

The strong variation of the mode shape during the very long settling time indicates a very slow electron capture and emission rate from the donor site. Comparing with the settling time during the temperature variation we find time constants for both emission and capture in the order of 2 -- 3 h.

Considering the small binding energy

- [4] M.V. Klein in *Light Scattering in Solids I*. Ed. M. Cardona , Topics in Applied Physics, Vol. 8 (Springer Berlin, 1983). p. 159ff
- [5] T. Kozawa, T. Kachi, H. Kano, Y. Taga, M. Hashimoto, N. Koide, K. Manabe, J. Appl. Phys. **75**, 1098 (1994).
- [6] C. Wetzel, W. Walukiewicz, E.E. Haller, J. Ager, I. Grzegory, S. Porowski, T. Suski, Phys. Rev. B **53**, 1322 (1996).
- [7] C.A. Arguello, D.L. Rousseau, S.P.S. Porto, Phys. Rev. **181**, 1351 (1969).
- [8] D.T. Hon, W.L. Faust, Appl. Phys, **1**, 241 (1973).
- [9] C. Wetzel, *Thesis*, Technical University Munich (1994).

