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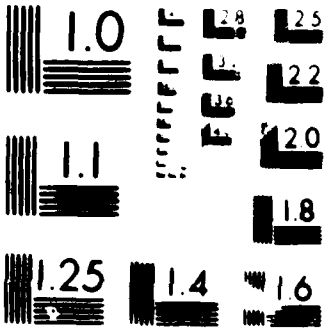
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REPORT DOCUMENTATION PAGE

Unclassitied	1b. RESTRICTIVE MARKINGS
2a SECURITY CLASSIFICATION AUTHORITY	3. DISTRIBUTION/AVAILABILITY OF REPORT: Approved for public release - distribution unlimited
2b DECLASSIFICATION/DOWNGRADING SCHEDULE	
4 PERFORMING ORGANIZATION REPORT NUMBER(S)	5. MONITORING ORGANIZATION REPORT NUMBER(S): AFOSR-TR- 87-1925
6a NAME OF PERFORMING ORGANIZATION: JILA	6b. OFFICE SYMBOL (If applicable):
6c ADDRESS (City, State and ZIP Code): University of Colorado, Campus Box 440 Boulder, Colorado 80309	7a. NAME OF MONITORING ORGANIZATION: Air Force Office of Scientific Research/NE
	7b. ADDRESS (City, State and ZIP Code): Bldg. 410, Bolling Air Force Base, Washington, DC 20332
8a NAME OF FUNDING/SPONSORING ORGANIZATION: Air Force Office of Scientific Research	8b. OFFICE SYMBOL (If applicable): NE
	9. PROCUREMENT INSTRUMENT IDENTIFICATION NUMBER: AFOSR-87-0119
8c ADDRESS (City, State and ZIP Code): Bldg. 410, Bolling Air Force Base, Washington, DC 20332	10. SOURCE OF FUNDING NOS.

PROGRAM ELEMENT NO.	PROJECT NO.	TASK NO.	WORK UNIT NO.
61102F	2305	K4	7

11 TITLE (Include Security Classification): Laser measurements of State-Resolved Ga and In Atom Sticking and Desorption on Metal and Semiconductor Surfaces (unclassified)

12 PERSONAL AUTHOR(S): Stephen R. Leone

13a TYPE OF REPORT	13b. TIME COVERED	14. DATE OF REPORT (Yr., Mo., Day)	15. PAGE COUNT
Technical Report	FROM 1/Dec/86 TO 30/Nov/87	87 December 1	8

16 SUPPLEMENTARY NOTATION

17 COSATI CODES			18. SUBJECT TERMS (Continue on reverse if necessary and identify by block number)
FIELD	GROUP	SUB GR	Semiconductors, GaAs, surfaces, lasers, Gallium Arsenide

19. ABSTRACT (Continue on reverse if necessary and identify by block number)

Work is carried out on the dynamics of Ga and As scattering, sticking, and desorption from silicon single crystals using laser probing of the Ga and As (dimer) gas phase species. In the last six months, results have been obtained for the binding energy of Ga on silicon. Structural patterns of Ga on silicon at various coverages have been determined by LEED studies. Results have been obtained for the desorption of two different Ga spin-orbit states and a model developed to explain the observed behavior. The desorption pre-exponential factors suggest a one-dimensional mobility of Ga on silicon. These results are relevant to the epitaxial growth of GaAs on silicon.

20 DISTRIBUTION/AVAILABILITY OF ABSTRACT	21. ABSTRACT SECURITY CLASSIFICATION
UNCLASSIFIED/UNLIMITED ☒ SAME AS RPT. ☐ DTIC USERS ☐	Unclassified

22a NAME OF RESPONSIBLE INDIVIDUAL	22b. TELEPHONE NUMBER (Include Area Code)	22c. OFFICE SYMBOL
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DD FORM 1473, 83 APR — EDITION OF 1 JAN 73 IS OBSOLETE. — UNCLASSIFIED — SECURITY CLASSIFICATION OF THIS PAGE

Annual Report (1 December 1987)

AFOSR-87-0119 (Stephen R. Leone)

Research Objectives

Work is continuing on the investigation of Ga and As desorption, scattering, sticking, and epitaxial growth on well-characterized silicon (100) single crystals. Laser, state-resolved probing is utilized to detect the individual states of the atoms and molecules of the gas phase species. In the last six months, measurements have been made on the following: (1) the binding energy of Ga on Si(100) by isothermal and temperature programmed desorption under conditions of varying coverage, (2) determination of the kinetic pre-exponential factor for Ga desorption, which indicates a one-dimensional mobility of the Ga on Si(100) at certain coverages, (3) investigation of the individual spin-orbit states in desorption, which show that a rapid equilibrium occurs between the spin-orbit states in the desorption process, (4) development of a model to describe accurately a general two-state desorption problem and to elucidate the influence of the second state on the desorption parameters, and (5) measurement of the Ga overlayer structures on Si(100) using LEED, which shows the patterns of structures and the ordered nature of the adsorption up to one monolayer. One paper is published, one more submitted, and several more in preparation.

Status of Research

Our approach is unique in that we use a laser to probe the densities of such species as Ga and As atoms and As_2 dimers as they scatter or desorb from the surface.

We have performed several kinds of experiments for Ga atoms interacting with a well-characterized Si(100) 2 × 1 reconstructed single crystal. In one kind of experiment, a Ga beam is exposed on the cold Si surface until a

A-1

certain coverage is achieved. The surface temperature is increased, and the laser probes the desorption of the Ga atoms. From these temperature programmed desorption studies we determine that the Ga desorbs with first order kinetics under low coverage. At higher coverages a zero order desorption occurs to lower temperature as Ga-Ga interactions on the surface start to be important.

The distinct point at which the monolayer changes over to a multilayer type of interaction can be correlated with the Auger and LEED data which we have taken. We find that the monolayer coverage corresponds to a zone from zero coverage to a coverage where all of the Si sites are occupied. Additional Ga then drastically changes the interaction. The LEED patterns (see below) show evidence that at coverages of about one monolayer, the Ga atoms most likely cause the Si, which is normally a double row structure of Si dimers, to open up and form a 1 × 1 pattern. The Auger data confirm that this is also the point when the Si surface is fully covered. We have observed all of the LEED structures up to one monolayer and have developed a model for the overlayer growth (Fig. 1). At low coverages, the Ga most likely occupies sites between the double rows of silicon atoms, forming bonds to two "dangling bonds" from silicon atoms and possibly dimerizing with neighboring Ga atoms. At high coverages of many monolayers, we have evidence for island formation. Specially stable structures occur at exactly one-half and one monolayer coverage. The persistent 8 × 1 pattern can be explained by the size of the covalent radii of Ga and Si, which are mismatched by just 7 Ga to 8 Si atoms.

Another type of experiment is used to make accurate determinations of the binding energy of the Ga atoms. The surface is left at a fixed temperature and is exposed to the Ga beam. The isothermal desorption of Ga atoms under

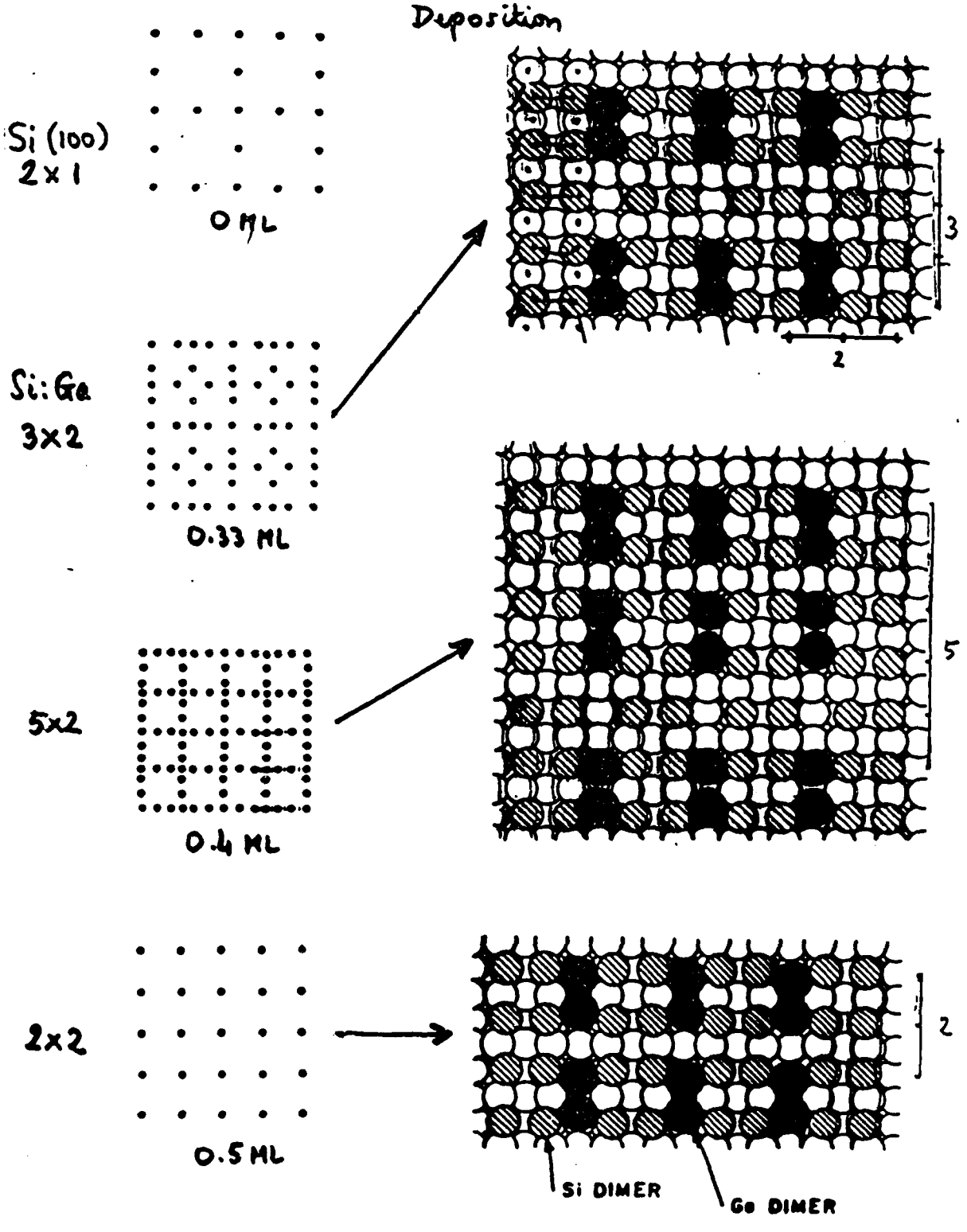
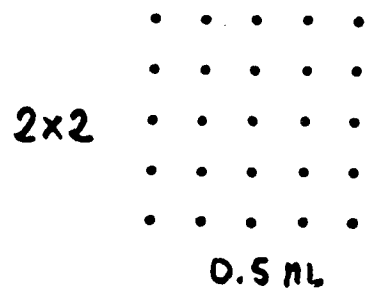
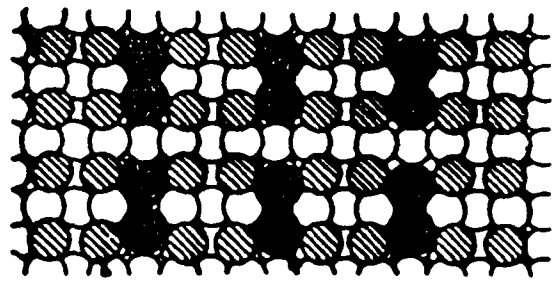
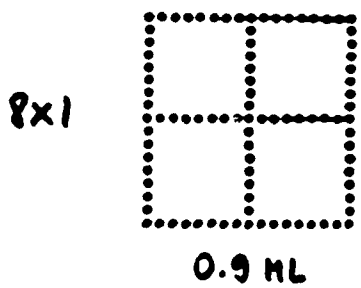

Deposition
Si (100)
2 x 1
0 ML
Si:Ga
3x2
0.33 ML
5x2
0.4 ML
2x2
0.5 ML
3
2
5
2
Si DIMER
Ga DIMER

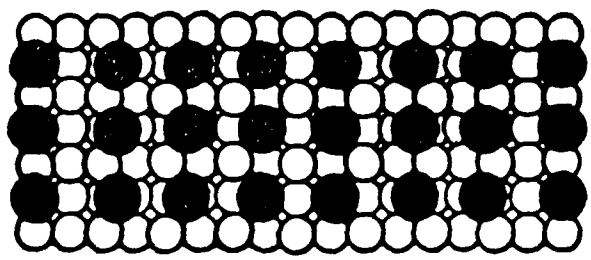
Figure 1

Deposition.

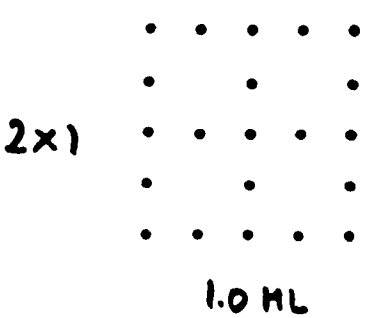








lateral bonding must be dominant
("metallic" VS "covalent")



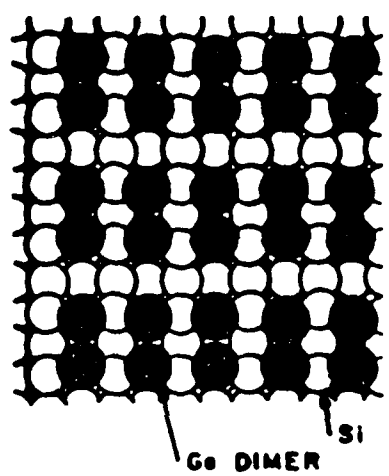


Figure 1 (continued)

low coverage is then measured with the laser by stopping the beam with a flag and watching the decay of the flux. A series of measurements are taken at different surface temperatures and an Arrhenius plot is made to extract both the desorption energy and the pre-exponential factor. Our results find that Ga is bound to Si(100) by 66 kcal $mole^{-1}$, or 2.9 eV (K. Carleton). The observed pre-exponential factor is 6×10^{14} s^{-1}. The strong binding energy has not been anticipated and has important consequences as a reason why both Ga and As can bind initially to the Si surface, thereby creating antiphase domains.

The pre-exponential factor is perhaps an even more exciting result. If an atom is completely mobile on the surface, then the desorption to the gas phase gains one more degree of freedom and leads to a theoretical pre-exponential factor of kT/h, or 2×10^{13} s^{-1}. If the atom is rigidly bound to one site on the surface (immobile), then the pre-exponential factor is 1×10^{16} s^{-1}. Our value gives strong evidence that the Ga atom is mobile only in one dimension on the Si(100) 2×1 surface at low coverages. An explanation is that the atom is mobile along the double rows of the silicon atoms, but cannot cross between the rows. Such a result is important in the epitaxial growth, where atoms have to be mobile enough on the surface to locate the growing edge site and bind there.

The binding energy has also been measured at higher coverages and is observed to change dramatically at special points. For the region of coverage between 0.5 and 1 monolayer, the binding energy is 2.3 eV and the pre-exponential factor changes to $8 \times 10^{12\pm1.2}$ s^{-1}. For very high coverages where islands, or liquid drops, are suspected, the zero order kinetics gives a binding energy of 2.6 eV.

Figure 2 shows the Ga:Si Auger peak ratio as a function of deposition and temperature. The sharp breaks are evidence for the formation of Ga islands above a certain coverage. At room temperature, the Ga can be deposited in uniform layers indefinitely. However, at 60°C, it can be seen that islands begin to form above 2.5 monolayers. At 330°C, islands form above 1 monolayer.

By correlating the LEED structures with the measurements of the binding energies, a complete picture of the Ga binding to Si has been achieved. It should be noted that our results differ from those of L. Feldman at AT&T Laboratories, in that he suggests the fundamental Ga binding energy is 2.2 eV. We find it is 2.9 eV at low coverages and 2.3 eV at ~0.5-1.0 monolayer. We believe that our much more careful analysis and the correlation to the LEED structures will stand the test of time. This work is in preparation for Surface Science (2 papers B. Bourguignon, R. Smilgys, K. Carleton).

Other results from our studies have found that a portion of the Ga goes into the bulk of the silicon and can be driven out only by very high temperatures. This state is currently under further investigation. In addition, from the isothermal scattering experiments, the sticking coefficient of Ga on Si is measured to be between 0.9 and unity at typical growth temperatures. We plan to make further measurements of this parameter.

Finally, we have explored the difference in binding of the two spin orbit states of Ga. Surprisingly, the two states desorb with identical energies. This result prompted us to consider the dynamics of a two state atom when it leaves the surface. The results we observe are consistent with a model in which the two spin orbit states are in equilibrium at the last moment before desorption. The 2.5 kcal $mole^{-1}$ splitting of the spin orbit states causes the lower state ($^2P_{1/2}$) to desorb with a higher rate. The depletion of this level is repopulated by the excess population in the upper $^2P_{3/2}$ state. Thus the

(B) AES and LEED VS Deposition time

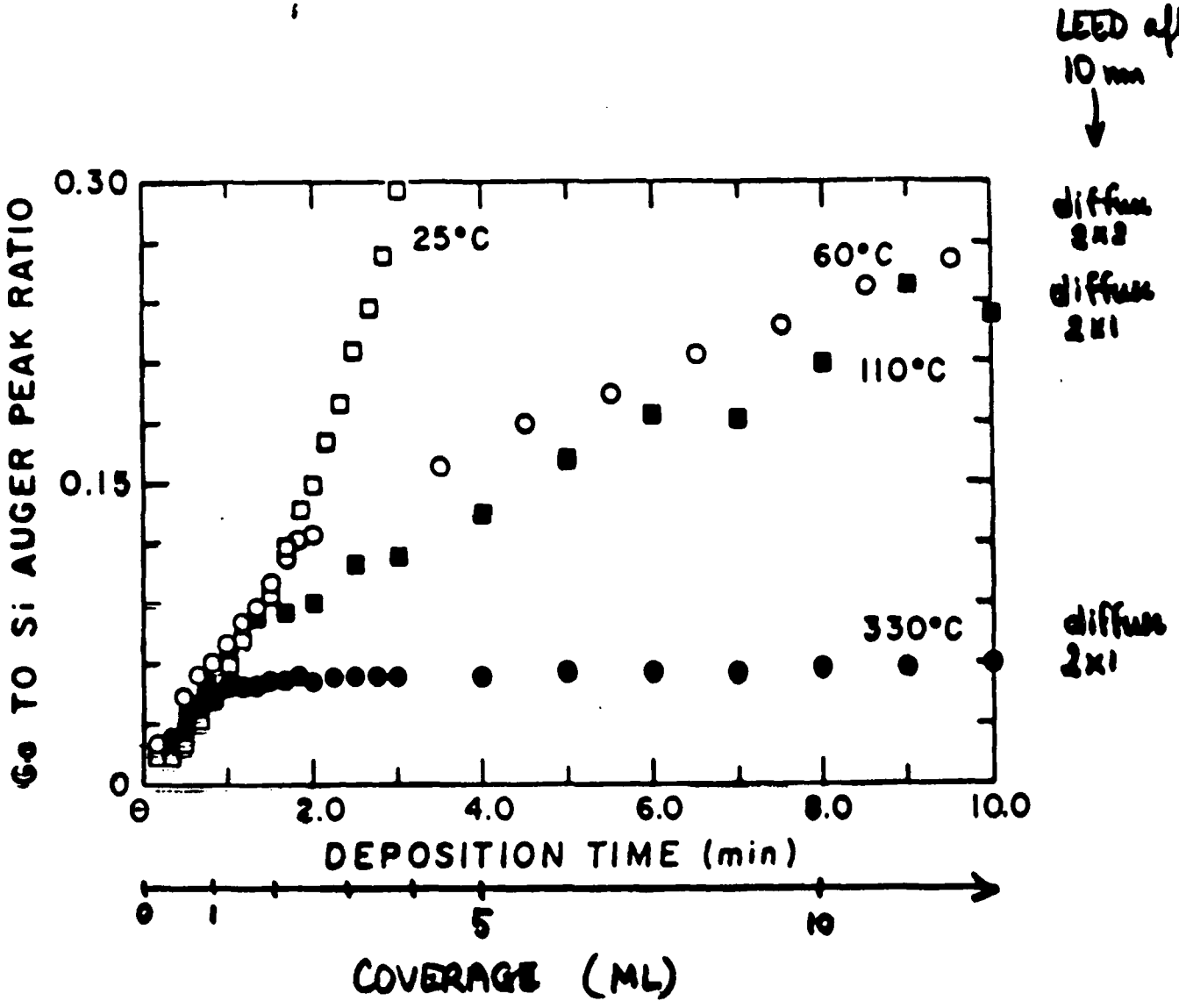


Figure 2

two atoms are "coupled" together in terms of their desorption dynamics. The population ratio, however, is a sensitive function of the energy of the splitting and any barriers to desorption for one or both atoms. This work has been submitted to Surface Science (K. Carleton, B. Bourguignon).

Professional Interactions

Talks

(S. Leone)

"Laser probing of gallium atom interactions with Silicon(100) surfaces," 14th Conference on the Physics and Chemistry of Semiconductor Interfaces, Jan. 1987 Salt Lake City, Utah.

"Laser probing of state-selected dynamics in the gas phase and on surfaces," University of California, Santa Barbara, California, April 1987.

(K. Carleton)

"Laser probing of gallium spin orbit state desorption and scattering from silicon(100) surfaces," Industry - University Advanced Materials Conference, Denver, Colorado, February 1987.

"Laser probing of gallium atom interactions with silicon," Colorado State University, Fort Collins, Colorado, December 1987.

(B. Bourguignon)

"Spin-orbit state selected detection of gallium desorbing from Si(100)," Gordon Conf. on the Dynamics of Gas-Surface Interactions (poster), August 1987, New Hampshire

Cumulative List of Publications

K. L. Carleton and S. R. Leone, "Laser probing of gallium spin orbit state desorption and scattering from silicon(100) surfaces," Proc. of the Industry - University Advanced Materials Conference, Denver 1987, ed. by J. G. Morse (The Metalurgical Society, Inc., Warrendale, PA, 1987), p. 9.

K. L. Carleton and S. R. Leone, "Laser probing of gallium atom interactions with silicon(100) surfaces," J. Vac. Sci. Technol. B5, 1141 (1987).

K. L. Carleton, B. Bourguignon, and S. R. Leone, "Desorption of a two-state system: Laser probing of gallium atom spin-orbit states from silicon(100)," submitted Surf. Sci,

(In preparation)
manuscript for European SASP conference.
manuscript on LEED structures of Ga on Si
manuscript on energetics as a function of coverage, Ga on Si

END

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