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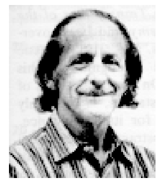

Current Comments®

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The 1985 Nobel Chemistry Prize to Jerome Karle and Herbert A. Hauptman and the Physics Prize to Klaus von Klitzing Contrast Delayed Versus "Instant" Recognition

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In last year's examination of the 1984 Nobel laureates in physics and chemistry,[1] I noted that Carlo Rubbia, Harvard University and the European Center for Nuclear Research (CERN), Geneva, Switzerland, and Simon van der Meer, also of CERN, received the physics prize barely two years after they identified the W and Z subatomic particles. This is one of the shortest intervals between discovery and recognition in Nobel history. By contrast, the 1984 winner in chemistry, R. Bruce Merrifield, Rockefeller University, New York, did the bulk of his prizewinning work in peptide synthesis in the 1960s.

A similar disparity is evident in the 1985 physics and chemistry prizes, which we'll be examining this week as we complete our discussion of last year's Nobel laureates. The physics prize was awarded for work done in the early 1980s. The chemistry prize, however, honored work that first appeared in the 1950s—work that found little recognition and acceptance when it was published.

Chemistry

The 1985 prize in chemistry was awarded to Herbert A. Hauptman, Medical Foundation of Buffalo, New York, and Jerome Karle, US Naval Research Laboratory, Washington, DC. Hauptman, a mathematician, and Karle, a physical chemist, both graduated from City College, New York, in the class of 1937—the same class that produced the 1959 Nobel laureate in medicine, Arthur Kornberg. In citing Hauptman and Karle, the Royal Swedish Academy of Sciences mentioned their "outstanding achievements in the development of direct methods for the determination of crystal structures."[2] Although these methods were not immediately accepted (as our citation data will demonstrate), they are widely applied today in research laboratories. Crystal structures that once took months to analyze can now be determined in a matter of hours using the methods developed by Hauptman and Karle.

Determining the structure of a molecule is essential to understanding its chemical bonding and its reactions and interactions with other molecules, according to Wayne A. Hendrickson, Department of Biochemistry and Molecular Biophysics, Columbia University College of Physicians and Surgeons, New York.[3] In order to design new drugs and synthesize rare natural products, it is important to have a precise, three-dimensional picture of the arrangement of atoms within the molecule. One of the most accurate methods for determining structures is by analyzing X-ray diffraction data from crystals, a method known as X-ray crystallography. X rays beamed through a crystal of a substance will be scattered by the atoms and molecules, producing a diffraction pattern that appears on photographic film as a cluster of dots of varying intensity. Information on the large-scale crystal geometry and the individual crystal units can be inferred from the intensities and phases of the scattered waves in the pattern.[4]

It is relatively easy to deduce the atomic structure of very simple crystals by means of X-ray crystallography. In order to solve more complicated structures, however, it is necessary to overcome the so-called "phase problem." As Hendrickson explains, the scattering of X-ray waves by crystals is restricted to certain discrete directions governed by the crystal lattice. Each of the diffracted waves is made up from the scattering by all atoms, and in complex molecules there may be hundreds of atoms. This complicated diffraction interference leads to a distinctive phase and magnitude for each allowed direction.[5] Measuring the intensity of the deflected X rays presents no problems, but determining the phase—that is, how much the waves in the various rays are displaced in relation to each other—is another matter. This phase information, essential in obtaining the molecular structure of a crystal from the X-ray diffraction pattern, was once thought to be irretrievable.

Hauptman and Karle, then working together at the US Naval Research Laboratory, published their "direct methods" in papers that appeared primarily between 1950 and 1956. Their statistical procedures permit researchers to extract the phase information and to compute the molecular structure directly from the data in the diffraction pattern. Many of these papers appeared in *Acta Crystallographica*. Their key publication from this period, however, was a 1953 monograph entitled *Solution of the phase problem. I. The centrosymmetric crystal.*[6] This work proposed the use of probability theory as a means of solving the phase problem. The methods described require highly complex mathematics. Consequently, this primordial work met with considerable resistance and neglect when first published. In fact, the direct methods were not accepted for more than a decade. The delayed recognition of this paper is demon-

Figure 1: Chronological distribution of citations from the *SCI*® to Hauptman and Karle's highly cited 1953 monograph, *Solution of the phase problem. I. The centrosymmetric crystal.*

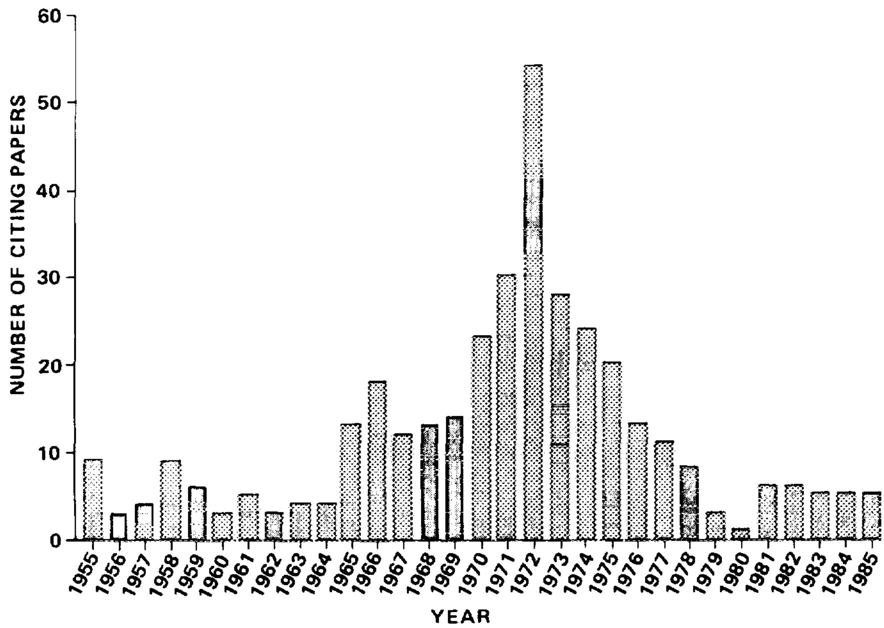


Table 1: Papers coauthored by Karle and Hauptman most cited in the *SCI*® from 1955 to 1985. The papers are arranged in descending order according to number of citations. A = total number of citations. B = bibliographic information. *SCI*® research fronts for which the paper is core are given in parentheses after the bibliographic information.

A	B
447	**Karle J & Hauptman H.** A theory of phase determination for the four types of non-centrosymmetric space groups 1*P*222, 2*P*22, 3$P_1$2, 3$P_2$2. *Acta Crystallogr.* 9:635-51, 1956. (73-1253, 72-0579, 71-1024, 70-0937)
363	**Hauptman H & Karle J.** *Solution of the phase problem. I. The centrosymmetric crystal.* New York: American Crystallographic Association, 1953. 87 p.
116	**Karle J & Hauptman H.** The phases and magnitudes of the structure factors. *Acta Crystallogr.* 3:181-7, 1950.
100	**Hauptman H & Karle J.** Structure invariants and semivariants for non-centrosymmetric space groups. *Acta Crystallogr.* 9:45-55, 1956. (72-0579)
97	**Karle I L, Hauptman H, Karle J & Wing A B.** Crystal and molecular structure of *p, p'*-dimethoxybenzophenone by the direct probability method. *Acta Crystallogr.* 11:257-63, 1958.
75	**Karle J & Hauptman H.** Application of statistical methods to the naphthalene structure. *Acta Crystallogr.* 6:473-6, 1953.

strated by its citation record. The graph in Figure 1 shows that it received fewer than 10 citations per year in the first 10 years or so after publication. Several successes in solving crystal structures, as well as advances in the high-speed computer technology necessary to perform the calculations, led to increasing acceptance of the technique by the end of the 1960s.[7] Our data indicate that this work has now been cited over 350 times. Whether this qualifies as a classic case of delayed recognition[8] is determined by one's reference point: the first 5 to 10 years after publication or the delay in recognition that is typically displayed by the Nobel committees.

Table 1 is a list of most-cited papers coauthored by Karle and Hauptman according to the *Science Citation Index*® (*SCI*®), 1955-1985. Their most-cited work is a 1956 paper from *Acta Crystallographica* entitled "A theory of phase determination for the four types of noncentrosymmetric space groups 1*P*222, 2*P*22, 3$P_1$2, 3$P_2$2."[9] This paper, cited over 400 times, expands on their 1953 monograph and discusses joint probability distributions and formulas to determine the phase of typical noncentrosymmetric space groups. In addition to the 1953 monograph,[6] Table 1 includes a 1950 paper from *Acta Crystallographica* on "The phases and magnitudes of the structure factors."[10] This work has been cited in over 100 publications.

Today, the Hauptman-Karle methodology is indispensable for determining the molecular structure of a variety of useful substances, including antibiotics, catalysts, alloys, and natural products such as peptides, steroids, and alkaloids.[11] Although direct methods have been applied to molecules containing up to 200 atoms, they are not yet powerful enough to be used with larger proteins. Both scientists, however, continue to work at extending the utility and power of the direct methods—Hauptman in Buffalo and Karle in Washington.

Hauptman

Herbert A. Hauptman was born in New York City in 1917. He received a BS in mathematics from City College, New York, an MA in mathematics from Columbia University, and a PhD in mathematics from the University of Maryland, College Park. He worked as a physicist and mathematician at the US Naval Research Laboratory from 1947 to 1970, before leaving to become executive vice president and research director of the Medical Foundation of Buffalo, where he has been ever since. Since 1970 he has also been a professor of biophysics at the State University of New York, Buffalo.

One of Hauptman's first papers, coauthored with Karle, was "The structure of atoms from diffraction studies," which appeared in *Physical Review* in 1950.[12]

Table 2: Chronological list of 1955-1985 *SCI*® citations to Karle and Brockway's 1944 paper in the *Journal of the American Chemical Society*. A = year. B = number of citations.

A	B	A	B	A	B
				1975	3
1955	4	1965	4	1976	2
1956	2	1966	4	1977	4
1957	7	1967	8	1978	1
1958	10	1968	8	1979	2
1959	14	1969	3	1980	3
1960	8	1970	7	1981	5
1961	6	1971	9	1982	2
1962	11	1972	5	1983	2
1963	6	1973	5	1984	2
1964	0	1974	7	1985	2

This paper, like the authors' later work, dealt with methods for obtaining the electron distribution in atoms from diffraction data.

Karle

Jerome Karle was also born in New York, in 1918. He received a BS in chemistry and biology at City College, New York, an MA in biology from Harvard, and MS and PhD degrees in physical chemistry from the University of Michigan, Ann Arbor. Since 1946 he has worked at the US Naval Research Laboratory, where he is now chief scientist of the Laboratory for the Structure of Matter.

Karle's first paper, "An electron diffraction investigation of the monomers and dimers of formic, acetic and trifluoroacetic acids and the dimer of deuterium acetate," coauthored with L.O. Brockway, Chemical Laboratory, University of Michigan, appeared in 1944.[13] This paper has been cited over 100 times. Table 2 shows year-by-year citations to this paper. In addition to the papers coauthored with Hauptman, Karle has coauthored several highly cited papers with his wife, Isabella, who is also a chemist at the US Naval Research Laboratory. One of these is a 1966 paper, "The symbolic addition procedure for phase determination for centrosymmetric and noncentrosymmetric crystals."[14] The work has been cited over 1,300 times. It was the subject of a *Citation Classic*® commentary in 1977, in which the authors noted that

> the analysis of crystal structures, facilitated by the symbolic addition procedure and the development of computers and automatic diffractometers, has had a major impact on the progress of many scientific areas. For example, the availability of such capabilities has played a significant role in the practice of organic chemistry by aiding in the identification of reaction intermediaries, final products, and the clarification of reaction mechanisms.[15]

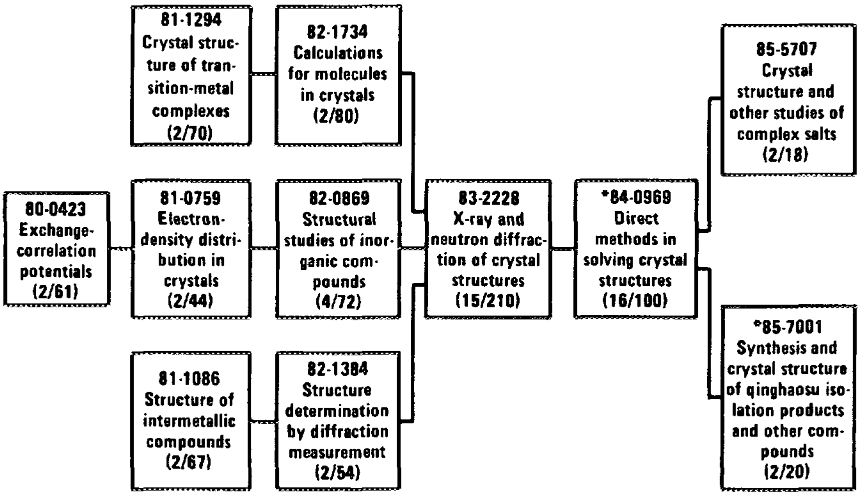
This paper has also been prominent in several of our citation studies over the years. These include a 1974 study of highly cited chemistry articles[16] as well as a study that same year of *Acta Crystallographica*.[17] It was also featured in a 1976 essay listing 50 highly cited articles from Scandinavian journals.[18] *Acta Crystallographica*, which appears in three separate sections covering various areas of crystallography, is published by Munksgaard, Copenhagen, Denmark.

Another highly cited paper coauthored by Isabella and Jerome Karle, with over 300 cites, describes the use of direct methods to determine the structure of the synthetic polypeptide cyclohexaglycyl.[19] The authors discussed this *Citation Classic* in 1980, noting that "there are a number of reasons why this publication has been highly cited. Not only had a simple and rapid procedure for phase determination been established but it was applied to a crystal of a substance with unusual features."[20]

Research-Front Data

Not surprisingly, many of the foregoing articles are identified with research fronts on crystallography and other structural and determinational techniques. One 1984 front is "Direct methods in solving crystal structures" (#84-0969). Karle's 1966 paper[14] and his 1968 article on "Partial structural information combined with the tangent for-

Figure 2: Historiograph of crystal-structure research. The numbers given in parentheses following the research-front titles refer to the number of core/citing items for each research front. Asterisks (*) next to the research-front numbers indicate research fronts in which Karle or Hauptman are core authors.



mula for noncentrosymmetric crystals"[21] are among the 16 core papers identified with this topic, on which about a hundred citing articles were published in 1984. Another 1984 front, "Crystal structure of crambin and other compounds using anomalous dispersion data or isomorphous replacement with direct methods" (#84-7162), includes five core publications, one of which, a 1982 paper by Hauptman from *Acta Crystallographica Section A*, is "On integrating the techniques of direct methods with anomalous dispersion. I. The theoretical basis."[22] Another core paper in this group is Karle's 1980 paper in *International Journal of Quantum Chemistry* on "Some developments in anomalous dispersion for the structural investigation of macromolecular systems in biology."[23]

Figure 2 is a historiograph, a string of annual research fronts, on crystal-structure research. As can be seen, research front #84-0969 continued into two 1985 fronts. In one of these, "Synthesis and crystal structure of qinghaosu isolation products and other compounds" (#85-7001) (qinghaosu is a plant extract used in treating malaria), the two core papers involved were mentioned above in connection with front #84-0969.[14,21]

Physics

In contrast to the 30 years that elapsed before the Karle-Hauptman work was recognized by the Nobel committee, the winner in physics received almost "instant" recognition.

The 1985 Nobel Prize in physics was awarded to Klaus von Klitzing, Max Planck Institute for Solid State Research, Stuttgart, Federal Republic of Germany (FRG), for the discovery of the quantized Hall effect, which, in the words of the Royal Swedish Academy of Sciences, "has fundamental implications for physics. His discovery has opened up a new research field of great importance and relevance."[24] The effect, first observed by von Klitzing and his colleagues in a 1980 experiment, will have great utility in establishing standards for the measurement of electrical resistance and in determining the precise value of a fundamental constant known as the fine structure constant.[24]

The American physicist Edwin H. Hall noted in 1879 that when a conductor carrying an electric current is subjected to a perpendicular magnetic field, the moving electrons that make up the current experience a lateral force, causing them to accumulate on one side of the conductor. The voltage difference across the conductor is called the Hall voltage. A measurement known as the Hall resistance can be obtained by dividing the Hall voltage by the current along the conductor.[25]

Von Klitzing discovered that under certain conditions the Hall resistance does not vary continuously in response to changes in the magnetic field but instead becomes *quantized*. That is, the resistance changes in definite, discrete steps based on quantum numbers. Von Klitzing observed the effect in an experiment at the High Magnetic Field Laboratory, Grenoble, France, with colleagues Gerhard Dorda, Siemens Research Laboratory, Munich, FRG, and Michael Pepper, Cavendish Laboratory, Cambridge, UK, who developed the experimental samples used in the study.[26] The experiment had originally been designed to investigate the effects of impurities and other factors in semiconductor devices.[27]

For this experiment, von Klitzing used what is known as a metal-oxide semiconductor field-effect transistor, or MOSFET. In this device, electrons are drawn into an extremely thin surface layer between a metal and a semiconductor. If this layer is thin enough and if the semiconductor is cooled to a temperature near absolute zero, the electrons can be forced to move in only two dimensions. It is under these conditions that the quantized Hall effect is observed when a perpendicular magnetic field is applied to the sample.

Theorists, in discussing the Hall effect in two-dimensional systems, had concluded that very complicated deviations from the classical behavior could be expected. Furthermore, it was postulated that under special conditions and within certain approximations the Hall conductivity would be related to an integral number multiplied by e^2/h, a fundamental constant of physics in which e is the electron charge and h is Planck's constant. No one had expected, however, that the quantization rule would apply with the accuracy observed by von Klitzing. The Hall resistance was found to exhibit step-like plateaus in which the values corresponded to integral numbers with a very high degree of precision.

Since 1980 researchers have been kept busy investigating the theoretical and experimental questions raised by von Klitzing's discovery. An entirely new phenomenon was observed in a 1982 study in which D.C. Tsui and colleagues, AT&T Bell Laboratories, Murray Hill, New Jersey, discovered what is now called the *fractional quantized Hall effect*. The plateaus in this study, rather than corresponding to integers as von Klitzing observed, reflected fractional numbers multiplied by the constant e^2/h.[28] This effect is currently the subject of intense investigation by researchers. Von Klitzing, meanwhile, continues to research aspects of two-dimensional electron systems and to explore the use of the quantized Hall effect as a standard of electrical resistance.[29]

Von Klitzing

Klaus von Klitzing was born in 1943 in Schroda in the region of Posen, a part of Poland that was annexed to Germany during World War II. His family moved to the FRG at the end of the war. He studied at the Technical University, Braunschweig, graduating in 1969, and received a doctorate in physics from the University of Würzburg in 1972. At the time of his prizewinning discovery in 1980 he was a teaching fellow at Würzburg. Since then he has also taught at the Technical University of Munich. He was named a director of the Max Planck In-

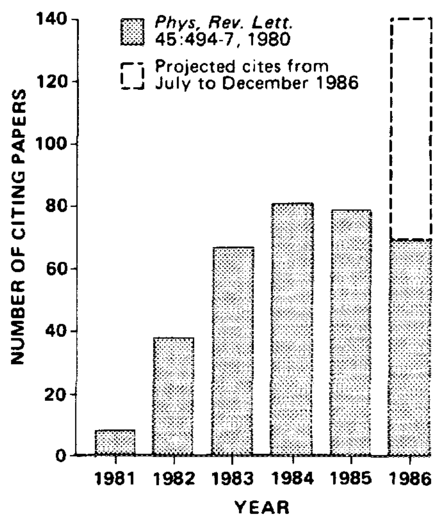
Table 3: Von Klitzing's papers most cited in the *SCI®* from 1955 to 1985. The papers are arranged in descending order according to number of citations. A = total number of citations. B = bibliographic information. *SCI* research fronts for which the paper is core are given in parentheses after the bibliographic information.

A	B
331	**von Klitzing K, Dorda G & Pepper M.** New method for high-accuracy determination of the fine-structure constant based on quantized Hall resistance. *Phys. Rev. Lett.* 45:494-7, 1980. (85-0102, 84-0099, 83-0897, 82-0541)
61	**Neugebauer T, von Klitzing K, Landwehr G & Dorda G.** Surface quantum oscillations in (110) and (111) *n*-type silicon inversion layers. *Solid State Commun.* 17:295-300, 1975.
38	**Bangert E, von Klitzing K & Landwehr G.** Self consistent calculations of electric subbands in p-type silicon inversion layers. (Pilkuhn M H, ed.) *Proceedings of the Twelfth International Conference on the Physics of Semiconductors.* 15-19 July 1974, Stuttgart, FRG. Stuttgart: Teubner, 1974. p. 714-8.

stitute for Solid State Research at the beginning of 1985.

Von Klitzing's first paper, coauthored in 1971 with G. Landwehr, University of Würzburg, dealt with the electrical properties of tellurium in strong magnetic fields.[30] Table 3 is a list of von Klitzing's most-cited works from the 1955-1985 *SCI*. Not surprisingly, perhaps, his most-cited work is the 1980 paper announcing the discovery of the quantized Hall effect.[26] It has been cited in over 300 papers. According to our data, no other paper published in *Physics Review Letters* in or since 1980 has achieved this popularity. As such, this work is a *Citation Classic*. The next most-cited work, "Surface quantum oscillations in (110) and (111) *n*-type silicon inversion layers," coauthored in 1975 with colleagues T. Neugebauer, University of Würzburg, Dorda, and Landwehr,[31] has been cited approximately 60 times, which is not unusual for articles published in *Solid State Communications*. Another paper, "Analysis of ϱ_{xx} minima in surface quantum oscillations on (100) n-type silicon inversion layers," coauthored with Würzburg colleague T. Englert in 1978,[32] has been cited approximately 25 times. In his Nobel lecture, von Klitzing noted that data indicating the quantized Hall effect are actually visible in this 1978 paper. However, because the data were analyzed on the basis of an incorrect model, the effect went unexplained at that time.[33] This may account for this paper's lower impact.

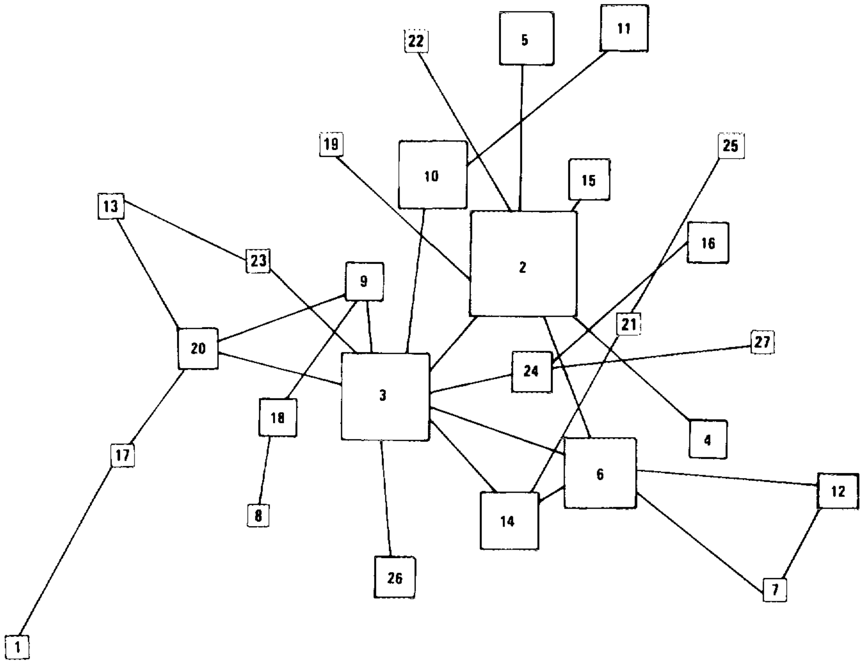
Figure 3: Chronological distribution of citations from the *SCI®* to von Klitzing's 1980 *Physical Review Letters* paper.



Research-Front Data

As the graph in Figure 3 indicates, von Klitzing's most-cited work, the 1980 paper from *Physical Review Letters*,[26] showed a distinct rise in citations in the four years after publication. It received approximately 70 citations in 1983, for example, and about 80 citations in both 1984 and 1985. Thus far in 1986 it has been cited in approximately 70 papers. It is 1 of 54 core papers in the research front "Electron localization and quantum transport phenomena in disordered electronic systems" (#85-0102). There were over 650 papers that cited into this

Figure 4: Multidimensional-scaling map for C2-level research front #85-0035, "Preparation and properties of semiconductors and transition metal compounds," showing links between C1-level research fronts. A = 1985 research-front number. B = research-front name. The numbers of core/citing items are given in parentheses following the research-front name.

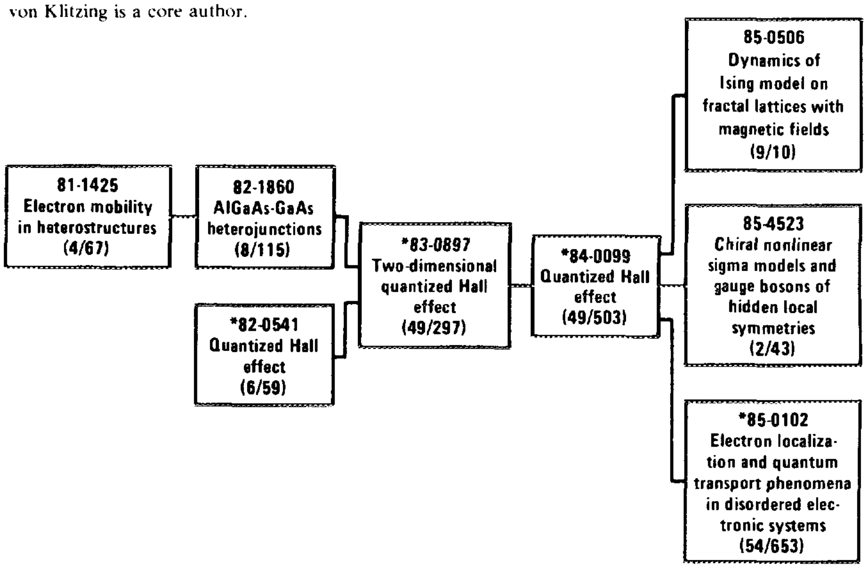


KEY

	A	B
1	0054	Theoretical and experimental study of amorphous semiconductor superlattices (2/28)
2	0102	Electron localization and quantum transport phenomena in disordered electronic systems (54/653)
3	0212	Energy bands in quantum well heterostructures and heterojunctions in semiconductors (36/485)
4	0859	Anderson localization and other properties of disordered or dirty anisotropic superconductors (4/51)
5	0864	Density of states and Anderson localization for one-dimensional Schrodinger operators with random potentials in disordered systems (10/98)
6	0908	Quantum transport phenomena in two-dimensional semiconductors (7/255)
7	0909	Impurities, conductivity, and other properties of silicon metal-oxide semiconductor systems (2/13)
8	1448	Quantum wells in doped superlattice structures (2/17)
9	1915	Impurity states in semiconductor quantum well structures by various methods (4/55)
10	1916	Photoluminescence and other studies of the properties of CDTE and GaAs superlattices, semiconductor quantum wells, and other heterostructures (28/246)
11	2598	Electronic structure and magneto-optical studies of holes in semiconductors (10/92)
12	3666	Electronic excitations in semiconductor superlattices (5/70)
13	3914	Resonant tunneling semiconductor heterostructures and their applications in devices (4/47)
14	4059	Carrier dynamics in quantum well structures using ultrafast laser pulses (13/110)
15	4096	Deep levels and electron traps in doped GaAs superlattice structures (7/64)
16	4252	Diffusion-induced disordering of GaAs multiquantum well lasers and superlattices during molecular beam epitaxial growth (10/62)
17	4909	Radiative recombination and design of semiconductor GaAs doping superlattices (4/45)
18	4987	Optical and other studies of quantum well systems grown by molecular beam epitaxy (3/38)
19	5129	Conductivity and localization theory of metallic surfaces (2/28)
20	5282	Bloch oscillations and structures of semiconductor superlattices and heterostructures (3/79)
21	5977	Quantum wells and photoluminescence in superlattices (2/36)
22	6037	Two-dimensional electron gas (4/33)

23 6503 Hot electron transport in selectively doped semiconductors (2/24)
24 6615 Molecular beam epitaxial growth of semiconductors using quantum well lasers (4/78)
25 7045 Preparation methods, photoluminescent properties, and electron microscopy studies of GaAs-GaAlAs multiquantum wells grown by organometallic chemical vapor deposition and molecular beam epitaxy (2/21)
26 8035 Semiconducting quantum well wires (2/15)
27 8391 Quantum well lasers and molecular beam epitaxy (2/27)

Figure 5: Historiograph of research on quantized Hall effect and two-dimensional electron systems. The numbers given in parentheses following the research-front titles refer to the number of core/citing items for each research front. Asterisks (*) next to the research-front numbers indicate research fronts in which von Klitzing is a core author.



front in 1985. This is indicative of the high level of research activity in this area.

The relationship between this and other 1985 fronts is demonstrated in Figure 4. This map shows the higher-level research front "Preparation and properties of semiconductors and transition metal compounds" (#85-0035). Front #85-0102 is also included in the historiograph in Figure 5, which shows the progression of annual research fronts on the quantized Hall effect and two-dimensional electron systems. As Harvard physicist Bertrand I. Halperin observes in his discussion of von Klitzing's work in *Science*, the discovery of the quantized Hall effect has led to a major revision of our understanding of electronic conduction in strong magnetic fields, and the subject remains very active for experimental and theoretical study.[29] H.-J. Czerwon, Scientific Information Center, Academy of Sciences of the German Democratic Republic, Berlin, and Jan Vlachý, Institute of Physics, Prague, Czechoslovakia, writing in the *Czechoslovak Journal of Physics*, have conducted an interesting follow-up study on publications and citations dealing with the quantized Hall effect in the wake of von Klitzing's Nobel Prize.[34]

This concludes our review of the 1985 Nobel laureates.

* * * * *

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