Spin-Polarized Electrons

In numerous experiments involving electrons, the ability to manipulate the spin of the electron gives unique additional information. For many years, however, such experiments were extraordinarily rare because of the absence of a good source of spin-polarized electrons. Up to the time this paper appeared, sources of spin-polarized electrons produced electron beams of very low intensity or polarization, or both, putting most polarized electron experiments out of reach and making those remaining heroic efforts. When GaAs Spin-Polarized Electron Source [1] appeared in print, a large number of proposed measurements were standing by, awaiting an improved source of spin-polarized electrons. The GaAs source proved to be the gateway to many fundamentally new types of experiments in atomic, condensed matter, nuclear, and particle physics.

The principle of the GaAs polarized electron source relies on 1) the photoexcitation of spin-polarized electrons in a solid and 2) their escape into vacuum. The origin of the spin polarization can be understood by referring to Fig. 1. GaAs is a direct-gap semiconductor with the band gap, E_{g} , at the center of the Brillouin zone as in the E(k) plot of the energy bands vs. crystal momentum k shown on the left side of Fig. 1. The relative intensities for transitions between m_i sublevels by photoexcitation with circularly polarized σ^+ and σ^- (positive and negative helicity) light are shown on the right side of Fig. 1. The polarization is defined as $P = (N_{\uparrow} - N_{\downarrow})/(N_{\uparrow} + N_{\downarrow})$ where $N_{\uparrow}(N_{\downarrow})$ are the number of electrons with spins parallel (antiparallel) to a quantization direction. Thus, for σ^+ light, quantum mechanical selection rules give the theoretical polarization, $P_{\rm th} = (1-3)/(1+3) = -0.5$ for band gap photoexcitation. An important characteristic of the GaAs source is that the sign of the spin polarization of the excited electrons can be easily changed by reversing the helicity of the incident light without affecting other parameters of the electron beam.



Fig. 1. On the left are depicted the energy bands of GaAs at the center of the Brillouin zone showing the band gap energy E_g and the spin-orbit splitting of the valence band Δ . On the right are shown the allowed transitions between m_j sublevels for circularly polarized light, σ^+ (solid lines) and σ^- (dashed lines), with relative transition probabilities given by the circled numbers.

Ordinarily, electrons excited to the conduction band minimum would be approximately 4 eV below the vacuum level and could not escape from the GaAs. However, by treating the surface of *p*-type GaAs with Cs and O_2 it is possible to lower the vacuum level at the surface below the energy of the conduction band minimum in the bulk to achieve the condition known as negative electron affinity (NEA) shown in Fig 2. NEA GaAs surfaces are extremely efficient photoemitters, which explains their widespread use in photomultiplier tubes, image intensifiers, and night vision devices. It is a lucky happenstance of nature that the world's best photoemitter is also an efficient source of spin-polarized electrons.



Fig. 2. GaAs surface activated with Cs and O₂ to achieve negative electron affinity E_A (vacuum level E_{∞} lower than the conduction-band minimum). Electrons excited across the band gap E_g by photons of energy $\hbar \omega$ thermalize to the conduction-band minimum, diffuse to the surface, and escape into the vacuum.

The purpose of the paper GaAs Spin-Polarized Electron Source was to describe how this effect, which had been discovered [2] a few years previously in spin-polarized photoemission experiments by Pierce and coworkers at the ETH-Zurich, could be used to provide a compact spin-polarized electron gun. GaAs Spin-Polarized Electron Source gives a comprehensive account not only of the theory of operation of this device, but also of its design, special materials preparation, construction, characterization, and performance. A crucial step is the cleaning of the GaAs photocathode surface, first by a series of chemical processes, and then by heating to just the right temperature in ultrahigh vacuum. The detailed descriptions of this and the sub-

sequent activation of the GaAs photocathode were important to enable others to build their own polarized electron guns. The analysis of the electron optics included the characteristics of the emitted beam, the cathode region, a 90° spherical deflector to change the longitudinal polarization of the emitted electrons to a polarization transverse to the electron momentum, and the transport and focusing system. The modular design provided a 1 keV electron beam suitable for transport through an isolation valve and on to the electron optics of a particular experiment. In this first application, the subsequent electron optics was designed to provide a collimated electron beam with variable energy from a few eV to a few hundred eV, as one would use in a variety of condensed matter physics experiments like spin-polarized low-energy electron diffraction (SPLEED).

The availability of an intense source of electrons with easily modulated degree of spin polarization had immediate impact. The two interactions, the spin-orbit interaction and the exchange interaction, that give rise to spin-dependent electron scattering from surfaces were investigated at NBS and given as examples of applications in the paper GaAs Spin-Polarized Electron Source. The spin-orbit interaction is due to the interaction of the spin of the electron with its own orbital angular momentum in scattering from a strong potential and is larger for materials of high atomic number. Large spin-dependent effects were observed in polarized electron scattering from a W(100) surface [3]. The spin dependent scattering asymmetry is readily determined by measuring the ratio of the part of the scattered intensity that varies with the modulation of the incident electron spin polarization to the part that is spin independent. Thus, the spin-dependent information is obtained from simple intensity measurements. The second example was scattering from a single-crystal Ni(110) surface where the spin dependence is due to the exchange interaction [4]. The exchange interaction is a consequence of the Pauli principle requiring the total wave function including spins to be asymmetric with respect to permutation of the particles. These polarized electron scattering measurements from a ferromagnetic surface pioneered a new, sensitive means to measure the degree of surface magnetic order. In another experiment [5] using the GaAs source, spin-polarized electron scattering from a ferromagnetic glass confirmed theoretical predictions that the temperature dependence of the surface magnetization at low temperatures should have the same power law as that of the bulk. However, the discovery of a larger prefactor than predicted led to recognition that the exchange coupling between the surface and the bulk is reduced.

In the 1980s, at NBS and in laboratories around the world, because of spin-polarized electron guns of the type described in GaAs Spin-Polarized Electron Source, researchers began making spin-polarized versions of their favorite electron spectroscopies. A case in point is spin-polarized inverse photoelectron spectroscopy (SPIPES). Inverse photoemission (IPES) is complementary to ordinary photoemission spectroscopy (PES). In particular, it permits investigation of unfilled states between the Fermi level and the vacuum level that are inaccessible in ordinary PES. Such states are crucial since it is the *d*-holes in transition-metal ferromagnets that are the "active ingredients" of the magnetism. SPIPES probes the spin-dependent nature of these states, in effect providing a magnetic spectroscopy of electron states. The first SPIPES measurements [6] were made on Ni(110) at NBS in a collaboration with Bell Laboratories colleagues who were involved in some of the first IPES measurements. Other spin polarized spectroscopies using the GaAs source were spinpolarized electron energy loss spectroscopy (SPEELS) [7] and spin-polarized low energy electron microscopy (SPLEEM) [8].

Among the longer term impacts of the paper GaAs Spin-Polarized Electron Source and the device described there is the use of such spin-polarizedelectron guns in the development of detectors of spinpolarized electrons. In early measurements at NBS, it was found that not only was the scattering of electrons from a magnetic surface spin dependent, but so also was the electron current absorbed in the target [9]. An electron spin analyzer was developed based on this effect [10]. While this spin analyzer turned out to be difficult to use, it was applied in a very important measurement of the spin polarization of the secondaryelectron energy distribution generated when an unpolarized electron beam is incident on a ferromagnet [11]. Energy- and spin-resolved measurements carried out at NBS showed that a ferromagnetic metal yields an abundance of highly polarized secondary electrons. This suggested the possibility of achieving high resolution imaging of magnetization of a surface by measuring the polarization of secondary electrons generated in a scanning electron microscope. This technique has come to be known as scanning electron microscopy with polarization analysis or SEMPA. The SEMPA magnetization image is formed by measuring the spin polarization of the secondary electrons as the SEM beam is "rastered" across the sample surface as shown schematically at the top of Fig. 3. The traditional Mott spinpolarization analyzer is large and heavy, in short cumbersome, and not suited for easy attachment to a scanning electron microscope. A new type of spin analyzer was developed at NBS, using the GaAs spinpolarized-electron gun to survey the phase space of materials, electron energies, and scattering conditions. This new low-energy diffuse-scattering spin analyzer [12] was fist-sized and at least as efficient as its big brother, the high-energy Mott analyzer. SEMPA has since been applied to numerous industrial problems by imaging domains in recording media, recording heads and other magnetic sensors, and magnetic randomaccess memory elements. The power of seeing the domains in high-resolution SEMPA images has proven very helpful in the development of magnetic randomaccess memory devices [13].

An especially important application of spin-polarized electrons is illustrated by a SEMPA investigation that took place in the early nineties [14]. At that time, there was a great deal of excitement within the condensed matter physics and magnetism communities as a consequence of a new discovery. A new kind of coupling was found to exist between magnetic layers separated by non-magnetic materials in very thin multilayer structures. The direction and extent of the coupling appeared to depend on the thickness of the non-magnetic layer in a way that did not correspond to any known theory and appeared not to depend strongly on the spacer material. Several innovations made the SEMPA investigation of this magnetic coupling unique. First, an Fe whisker was used as an atomically-perfect substrate. Second, the Cr spacer layer was grown in the shape of a wedge so that the variation of spacer thickness would be continuous as shown at the top of Fig. 3. Third, reflection high energy electron diffraction of the Cr wedge before the top Fe layer was deposited determined the average thickness at each point along the wedge to 0.1 atomic layer. Finally, SEMPA produced a single magnetization image of the top Fe layer, lower part of Fig. 3, where the magnetization changes back and forth from being parallel to the magnetization of the Fe whisker substrate (white regions) to antiparallel (black) as the Cr spacer thickness increases. Two superposed but distinct periods of oscillation of the exchange coupling with Cr thickness were observed. By varying the Cr growth temperature, it was possible to vary the roughness and show that rougher interfaces result from lower temperature growth. For such rough layers, the long-period coupling dominates as illustrated at the bottom right of Fig. 3. This explains why the short-period coupling had not been observed in the less perfect samples previously studied. Clearly, the universal period idea was shown to be invalid. The existence of the second period helped show the relationship of the coupling periodicity to the electronic structure of the spacer-layer material.



Fig. 3. In scanning electron microscopy with polarization analysis (SEMPA) the spin polarization of secondary electrons generated by a moving SEM beam is measured to obtain an image of the magnetization. Two such images of the magnetization of the top layer of a Fe/Cr (wedge)/Fe(001) sandwich are shown at the bottom of the figure for growth of the Cr wedge on the Fe whisker substrate at 200 °C and 300 °C. White (black) shows that the top Fe layer is coupled ferromagnetically (antiferromagnetically) to the Fe substrate.

Our discussion of the impact of *GaAs Spin-Polarized Electron Source* has so far been restricted to examples from condensed matter physics. At the time of publication of this paper, there were also experiments in the area of electron-atom collision physics just waiting for such a spin-polarized electron gun. When polarization techniques are used to fully state select the target and the incident electron polarization, then a complete or "perfect" scattering experiment is possible. In this case the quantum amplitude and phases can be measured instead of the cross-sections. Since cross-sections are sums of squares of complex amplitudes, direct determination of the individual amplitudes provides a more helpful comparison to, and a much more stringent test of, theoretical models. Using a GaAs polarized electron gun to scatter from a beam of Xe atoms, and measuring the change in polarization after scattering, the spin-orbit interaction was investigated in a complete experiment [15]. At NIST, the exchange interaction was carefully investigated in extensive experiments of elastic and inelastic polarized-electron scattering from state-selected Na beams [16].

GaAs Spin-Polarized Electron Source has been cited approximately 250 times, though nowadays the GaAs spin-polarized-electron gun is taken as accepted technology and its origins are often no longer cited. As discussed in a 1995 review [17], for most atomic and condensed matter experiments, the polarized electron gun described in GaAs Spin-Polarized Electron Source is more than adequate and remains the source of choice. GaAs polarized-electron sources for experiments in the areas of nuclear and particle physics, while working on the same principle, have been developed specifically to meet particular accelerator requirements such as a very high cathode voltage or a particular time structure of the beam. Advances over the years in GaAs type polarized electron sources, especially the progress toward higher polarization while maintaining sufficient beam intensity, can be attributed in large part to the demanding requirements on polarized electron sources for accelerator applications. Spin-polarized electrons play a crucial role in many particle physics experiments today.

The authors of *GaAs Spin-Polarized Electron Source* formed a strong team. The electron-optical design was led by Chris Kuyatt, assisted by Anija Galejs. Kuyatt was Section Chief of the Surface and Electron Physics Section at the time of the beginning of construction of the spin-polarized electron source. By the time the paper was published, he had become Chief of NIST's Center for Radiation Physics. In the 1960s, he and John Simpson developed high resolution electron monochromators and energy analyzers [18] and with them investigated sharp resonances in the electron transmission in gases [19]. The explanation of the resonance lineshapes is found in the landmark paper by Ugo Fano [20] (discussed elsewhere in this volume). Kuyatt worked at NIST until his death in 1998.

Robert Celotta, who became group leader after Kuyatt, and later a NIST Fellow, continues to do both research and serve as Group Leader of the Electron Physics Group. Dan Pierce also became a NIST Fellow and continues his research in the NIST Electron Physics Group. Together, Celotta and Pierce received the Edward Uhler Condon Award for distinguished achievement in written exposition as a result of their contribution to the paper GaAs Spin-Polarized Electron Source. They also shared the Department of Commerce Gold Medal and the American Vacuum Society Gaede-Langmuir Prize for their development of advanced, spin-polarized electron beam technology. Celotta and Pierce were given the first NIST William P. Slichter Award, which recognizes the growing need to promote cooperative partnerships between government and industrial researchers.

Stan Mielczarek and Anija Galejs worked as physicists in the Electron Physics Group until their retirements in 1990 and 1988 respectively. Bill Unertl was a National Research Council Postdoctoral Associate who left to take a faculty position at the University of Maine, where he is now Professor of Physics. Gwo-Ching Wang was a postdoctoral fellow and now is Professor of Physics at Rensselaer Polytechnic Institute. The National Bureau of Standards received IR-100 Awards in 1980 and 1985 (selected by Industrial Research and Development Magazine as one of the 100 most significant technical innovations of the year) for a Surface Magnetometer and for an Electron Gun, both of which were outgrowths of the development described in *GaAs Spin-Polarized Electron Source*.

Prepared by Daniel T. Pierce and Robert J. Celotta.

Bibliography

- D. T. Pierce, R. J. Celotta, G.-C. Wang, W. N. Unertl, A. Galejs, C. E. Kuyatt, and S. R. Mielczarek, GaAs Spin Polarized Electron Source, *Rev. Sci. Instrum.* 51, 478-499 (1980).
- [2] D. T. Pierce, F. Meier, and P. Zürcher, Negative Electron Affinity GaAs: A New Source of Spin-Polarized Electrons, *Appl. Phys. Lett.* 26, 670-672 (1975); D. T. Pierce and F. Meier, Photoemission of Spin-Polarized Electrons from GaAs, *Phys. Rev. B* 13, 5484-5500 (1976); D. T. Pierce, F. A. Meier, and H. C. Siegmann, *Source of Spin Polarized Electrons*, U.S. Patent 3,968,376, issued July 6, 1976.
- [3] G.-C. Wang, B. I. Dunlap, R. J. Celotta, and D. T. Pierce, Symmetry in Low-Energy Polarized-Electron Diffraction, *Phys. Rev. Lett.* 42, 1349-1352 (1979).
- [4] R. J. Celotta, D. T. Pierce, G.-C. Wang, S. D. Bader, and G. P. Felcher, Surface Magnetization of Ferromagnetic Ni(110): A Polarized Low-Energy Electron Diffraction Experiment, *Phys. Rev. Lett.* **43**, 728-731 (1979).
- [5] D. T. Pierce, R. J. Celotta, J. Unguris, and H. C. Siegmann, Spin-dependent Elastic Scattering of Electrons from a Ferromagnetic Glass, Ni₄₀Fe₄₀B₂₀, *Phys. Rev. B* 26, 2566-2574 (1982).
- [6] J. Unguris, A. Seiler, R. J. Celotta, D. T. Pierce, P. D. Johnson, and N. V. Smith, Spin-Polarized Inverse Photoelectron Spectroscopy of Solid Surfaces: Ni(110), *Phys. Rev. Lett.* 49, 1047-1050 (1982).
- [7] J. Kirschner, D. Rebenstorff, and H. Ibach, High-Resolution Spin-Polarized Electron-Energy-Loss Spectroscopy and the Stoner Excitation Spectrum in Nickel, *Phys. Rev. Lett.* **53**, 698-701 (1984); D. L. Abraham and H. Hopster, Spin-Polarized Electron-Energy-Loss Spectroscopy on Ni, *Phys. Rev. Lett.* **62**, 1157-1160 (1989).
- [8] E. Bauer, Low Energy Electron Microscopy, *Rep. Prog. Phys.* 57, 895-938 (1994).
- [9] H. C. Siegmann, D. T. Pierce, and R. J. Celotta, Spin-dependent Absorption of Electrons in a Ferromagnetic Metal, *Phys. Rev. Lett.* 46, 452-455 (1981).
- [10] R. J. Celotta, D. T. Pierce, H. C. Siegmann, and J. Unguris, An Electron Spin Polarization Detector: Spin-Dependent Absorption of a Polarized Electron Beam, *Appl. Phys. Lett.* 38, 577-579 (1981).
- [11] J. Unguris, D. T. Pierce, A. Galejs, and R. J. Celotta, Spin and Energy Analyzed Secondary Electron Emission from a Ferromagnet, *Phys. Rev. Lett.* **49**, 72-76 (1982).
- [12] J. Unguris, D. T. Pierce, and R. J. Celotta, Low-Energy Diffuse Scattering Electron-Spin Polarization Analyzer, *Rev. Sci. Instrum.* 57, 1314-1323 (1986).
- [13] J. Daughton, Non-volatile Electronics, private communication.
- [14] J. Unguris, R. J. Celotta, and D. T. Pierce, Observation of Two Different Oscillation Periods in the Exchange Coupling of Fe/Cr/ Fe(100), *Phys. Rev. Lett.* 67, 140-143 (1991).

- [15] W. Wübker, R. Möllenkamp, and J. Kessler, "Perfect" Elastic e⁻-Xe Scattering Experiment, *Phys. Rev. Lett.* **49**, 272-275 (1982).
- [16] J. J. McClelland, M. H. Kelley, and R. J. Celotta, Spin-Dependent Superelastic Scattering from Pure Angular Momentum States of Na(3P), *Phys. Rev. Lett.* **56**, 1362-1365 (1986); J. J. McClelland, M. H. Kelley, and R. J. Celotta, Superelastic Scattering of Spin-Polarized Electrons from Sodium, *Phys. Rev. A* **40**, 2321-2329 (1989); S. R. Lorentz, R. E. Scholten, J. J. McClelland, M. H. Kelley, and R. J. Celotta, Spin-Resolved Elastic Scattering of Electrons from Sodium, *Phys. Rev. A* **47**, 3000-3006 (1993).
- [17] D. T. Pierce, Spin-Polarized Electron Sources, in Atomic, Molecular, and Optical Physics: Charged Particles (Experimental Methods in the Physical Sciences 29A), F. B. Dunning and Randall G. Hulet (eds.), Academic Press, San Diego (1995) pp. 1-38.
- [18] C. E. Kuyatt and J. A. Simpson, Electron Monochromator Design, *Rev. Sci. Instrum.* 38, 103-111 (1967).
- [19] C. E. Kuyatt, S. R. Mielczarek, and J. A. Simpson, Energy Losses and Elastic Resonances in Electron Scattering from H₂, *Phys. Rev. Lett.* **12**, 293-295 (1964).
- [20] U. Fano, Effects of Configuration Interaction on Intensities and Phase Shifts, *Phys. Rev.* **124**, 1866-1878 (1961).