Comment on "Optically pumped spin-exchange polarized-electron source"

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M. Pirbhai *et al.* [Phys. Rev. A **88**, 060701(R) (2013)] reported a new optically pumped spin-exchange polarized-electron source developed to avoid the limitations of the traditional GaAs-type source and to enable a broader range of experiments. The spin-exchange gaseous source has introduced its own limitations with 650-mW laser power producing only 4 μA of spin-polarized current with a 24% polarization and a 2-eV energy spread depending markedly on tenuous roles of various buffer gases which we consider are not explained cogently. Spin-related beam experiments over many decades have significant achievements using GaAs spin sources.

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I. Introduction. The development of a new optically pumped spin-exchange polarized-electron source [1] was motivated to avoid difficulties with the traditional GaAs-type source and to enable a broader range of experiments. We address these aspects in turn.

II. The Gaseous Spin-Exchange Source. The new source introduces operating characteristics we consider are not explained convincingly and are not more manageable than those of a GaAs source. The spin production mechanism occurs when an unpolarized electron beam exchanges spin with optically pumped Rb atoms in the presence of a buffer gas. Much uncertainty arises from the multiple roles that the buffer gas plays in providing strong coupling between the optical pumping and the electron generation processes even though the authors present a viable scenario. Figure 2 of Ref. [1] shows that detuning the laser by about its linewidth of 2 GHz has the side effect of hyperfine spin reversal [2] and so changes the spin polarization from about +0.05 to −0.05%. How was it shown that the beam tuning remained constant throughout an experiment and did not change the degree of polarization of the beam and remain independent of reversal of the pump laser helicity? Even though the source is in its early stage of development some expectation of the way to control this effect on performance is desirable.

The authors state that calculations indicate that the buffer gas ethylene causes scattered electrons to quasielastically thermalize 100 times faster than in nitrogen and so increase the cross section for spin exchange. The dominant scattering processes to quasielastically thermalize the gaseous mixture needs clarification whereas in the same context "electron thermalization" is used with its usual meaning of energy-loss processes.

Radiation trapping was discussed for the relatively high pressures in the source only as a reduction in intensity of the beam rather than its angular dispersion and change in photon polarization. Since pressure-dependent decay rates are known to effect the orientation and alignment of excited atomic states via the Dyakonov-Perel mechanism [3,4], do they consequently introduce uncertainties into the vector polarization with limitations for the consistent and known operation of the source?

An electron optical figure of merit for an electron-beam source may be defined as the emittance phase space or the product of energy, area, and solid angle. The value for the spin-exchange source is not clear but needs to be indicated with its dependence on the emergent electron energy. In comparison, a good value for a GaAs source is on the order of $0.04\,\mathrm{eV\,mm^2\,sr^{-1}}$ although a lower current beam with smaller brightness can be useful when searching for scattering asymmetries.

In the gaseous spin-exchange source, the spin-polarized electron beam is transported in a magnetic field of 0.02 T (200 G) with unspecified gradients which would rotate the polarization vectors of very slow electrons. This limitation may be of most concern during a search for chirality which requires observations of either in-plane polarization or polarization-dependent beam attenuation or a rotation of polarization relative to the incident electron momentum. What is the magnitude of the perturbation of such observations to be expected from the source?

The manageability of a gaseous spin-exchange source is significant with the need for a powerful 650-mW laser type-3B laser for which extensive training and safety requirements are considerable compared with a GaAs 1-mW type-1 laser. What are the operating time and long term stability of the source as they are influenced by the amount of consumables of Rb and buffer gas ethylene required as well as the maintenance of the 650-mW laser power to produce a 4-μA beam current with an energy spread of about 2 eV with 24% spin polarization?

III. Range of Experiments. The significant progress in spin-polarized electron-scattering physics during the last 40 years, reviewed for example by Kessler [5] and Gay [6], was achieved in nearly all cases using a traditional GaAs spin source even though other usable spin-polarized sources based on field emission [7] and spin-exchange atoms [8] were available. Nevertheless it was claimed [1] that destruction of the photocathode's negative electron affinity surface conditions by organic and other vacuum contaminants can make GaAs sources unusable or, at best, highly problematic. Extensive evidence to the contrary is available from varied experimental environments and types of measurements. GaAs sources endured experimental environments ranging from ultraclean 3×10^{-11}-Torr UHV surface studies [9] to dynamic differential pumping for noncondensable inert gas atomic beams [10], condensable vapors of metallic atoms [11],

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and chiral organic molecules, such as camphor-lanthanoid complexes and bromomethylbutane [12]. A recent paper [13] reports considerable success observing an asymmetry less than about 10^{-4} for the chirally sensitive electron-induced breakup of a bromocamphor molecule and adds much support to the continued use of GaAs as the spin-polarized electron source.

The demands on any electron spin source include constant beam characteristics, particularly stability of the incident polarization vector in both magnitude and direction which set a standard for the gaseous exchange source to meet. Also the need for higher spin polarization is found from a very broad range of types of users and experiments exploring quantum scattering phenomena producing electron spin asymmetries. GaAs has been preferred for surface studies including the electronic structure, energy- and momentum-resolved exchange and spin-orbit interaction [14], ultrafast spin-dependent dynamics in ferromagnetic thin films [15], and two-electron pair spectroscopy of the influence of Ni layers on Co film on W(110) [16]. Similar sources were used for gaseous atom and molecule studies of spin scattering effects in structure and dynamics [13,17] and the topological angular momentum in electron exchange excitation of a single atom [4]. To summarize, these effects have varied over five orders of magnitude in low electron energy scattering probability and required changes in electron spin vector polarization, all of which used the excellent performance of GaAs (and GaAs-based superlattices) spin sources with polarization up to 70%.

Furthermore, the future use of GaAs, both as an electron spin source and as a precursor of new physics, was heralded by an extremely high brightness of 10^7-A cm^{-2} sr^{-1} spin current with 90% polarization achieved with two-photon laser photoemission from strained-superlattice GaAs/GaAsP structures [18]. To summarize, there is much evidence indicating that GaAs sources are neither unusable nor highly problematic.

In conclusion the introduction to the paper by Pirbhai *et al.* overstates difficulties with GaAs sources. The ability of a standard GaAs spin source to achieve and maintain fundamental beam characteristics of electron optical and polarization transport has been indicated for many physical phenomena covering spatial, time, or polarization correlations [1,13,19]. In particular the GaAs Pierce-type sources at Perth [20–23] usually operate continuously for several months, routinely, without attendance of any kind other than continuous fluxes of cesium onto the GaAs surface enabling near constant emission current and polarization for both strained and unstrained activated GaAs surfaces. The main single common factor of good performance GaAs sources is the maintenance of a pressure of about 3×10^{-11} Torr and preferably lower at the crystal surface, irrespective of vacuum system spatial configurations and target environment. We note that a commercial spin-polarized electron source of the standard GaAs type is available [24].

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