## New Spin-Polarized Electron Source Based on Alkali Antimonide Photocathode

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New spin-dependent photoemission properties of alkali antimonide semiconductor cathodes are predicted based on the detected optical spin orientation effect and DFT band structure calculations. Using these results, the  $Na_2KSb/Cs_3Sb$  heterostructure is designed as a spin-polarized electron source in combination with the  $Al_{0.11}Ga_{0.89}As$  target as a spin detector with spatial resolution. In the  $Na_2KSb/Cs_3Sb$  photocathode, spin-dependent photoemission properties were established through detection of a high degree of photoluminescence polarization and high polarization of the photoemitted electrons. It was found that the multi-alkali photocathode can provide electron beams with emittance very close to the limits imposed by the electron thermal energy. The vacuum tablet-type sources of spin-polarized electrons have been proposed for accelerators, which can exclude the construction of the photocathode growth chambers for photoinjectors.

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In recent decades, the physics of spin-polarized electrons in semiconductors has proven to be not only a fascinating area of basic research, but also a fruitful field of device applications that have had a significant impact on several areas of modern physics. Spin-polarized electron beams with high currents are required by modern and future nuclear physics facilities such as the Electron Ion Collider, the International Linear Collider [1–5] and for developing new generation of electron-positron collider "super charmtau factory" [6] with a variety of uses.

Long lifetime polarized electron sources are also of interest for electron microscopy methods exploiting spin polarization to probe magnetization in materials and nanostructures [7,8]. Probing the spin-resolved electron states of solids, surfaces, and nanostructures give direct access to phenomena like magnetism [9], proximity effects [10], spin-orbit interaction [11], and related spin texture in lowdimensional systems [12], which are regarded as potential functional systems for developing low-energy fast spintronic sensors or logic devices.

Almost all modern electron sources for highly spinpolarized electron beams in accelerator physics and electron microscopy rely on photocathode materials based on III-V (GaAs) technology. The first realization of a GaAs spin-polarized source was made by Pierce *et al.* [13] in 1975 based on both the phenomenon of optical orientation of electron spins in semiconductors (creation of spin-oriented carriers with absorption of circularly polarized light) and the discovery of negative electron affinity (NEA) activation of *p*-type semiconductor surfaces. The appearance of charge carriers with oriented spins upon absorption of a circularly polarized light was originally studied by Garvin *et al.* [14] and by Lampel and Weibush [15]. Despite the widespread use of GaAs as a photocathode, a serious disadvantage of the GaAs cathode is its sensitivity to residual vacuum gases and, as a consequence, a short lifetime [16]. A more stable semiconductor electron emitter sources are based on the alkali metal antimonide cathodes [4].

Alkali antimonide semiconducting materials are of great interest for single photon detection in photomultiplier devices [17–19] and as electron sources for the generation of high brightness electron beams for the next generation light sources like Energy Recovery Linacs and Free Electron Lasers [3,20,21]. The advantage of these photocathodes is provided by their faster temporal response compared to III-V photoemitters. The multi-alkali photocathode can provide bright electron beams with intrinsic emittance very close to the limits imposed by the electron thermal energy [22]. The epitaxial growth of thin films of the high-efficiency photocathode alkali antimonide materials by molecular-beam epitaxy opens the way to a significant increase in brightness and efficiency near the threshold by reducing surface disorder [23–25]. All this leads to the

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FIG. 1. Calculated bulk electronic band structures of (a)  $Na_2KSb$  and (b) GaAs. Gray lines show spectra calculated without SOI included. Colored dots show orbital character of the states (see keys).

fact that some groups switch from GaAs photocathode to alkaline antimony photocathode for dc gun or rf gun [1].

First attempts to use photosensitive materials, such as  $Cs_3Sb$ ,  $K_2CsSb$ ,  $Na_2KSb$ , and  $Na_2KSb$ : Cs for electron sources have been performed since the 1930s [26–28], however, surprisingly, the possibility of their use as a source of spin-polarized electrons has not been studied so far.

This Letter presents the first observation of spin-polarized photoemission from alkali-antimonide semiconductor heterostructures. The vacuum spin photodiode consisting of semiconductor Na<sub>2</sub>KSb photocathode and Al<sub>0.11</sub>Ga<sub>0.89</sub>As spin detector, both activated to NEA states, is implemented and high spin polarization of photoemitted electrons from Na<sub>2</sub>KSb measured in spectral and image modes is demonstrated.

From a technological point of view, knowledge of the electronic structure and spin texture is essential in order to understand the spin-polarized photoemission properties. The calculated dispersion of the electron bands of cubic Na<sub>2</sub>KSb is shown in Fig. 1(a). In the density functional theory (DFT) calculations performed with the Vienna *ab initio* simulation package (VASP) [29,30], where the generalized gradient approximation (GGA-PBE) [31] for the exchange-correlation potential was applied and the interaction between the ion cores and valence electrons was described by the projector augmented-wave method [32,33], the alkali metals *p* orbitals were treated as valence electrons. To obtain realistic bulk gap the Slater-type DFT-1/2 self-energy correction method [34,35] with a partially (quaternary) ionized antimony potential was applied.

At first glance, one can see a very similar band structure for  $Na_2KSb$  and GaAs [calculated within similar approach, Fig. 1(b)]. Gray lines in Fig. 1 show the spectra calculated with switched-off spin-orbit interaction (SOI) for both  $Na_2KSb$  and GaAs, where three topmost valence bands are degenerate at  $\Gamma$  and the band gaps are 1.58 and 1.62 eV, respectively. The calculated energy band dispersion of the nonrelativistic electronic structure of Na2KSb and symmetries of the bands in the center of the Brillouin zone agree well with the earlier calculations performed within the localized spherical wave method [36] with the exception of smaller band gap in the latter case (0.9 eV), as well as with results obtained within GW approximation (1.51 eV) [37] with which there is a fine agreement. Switching SOI on leads to a slight narrowing of the band gaps, to 1.41 and 1.51 eV, respectively. However, the main changes occur in the valence band, that is by 90% is determined by the p states of Sb(As). The heavy-hole and light-hole valence bands remain degenerate while the third band splits off. This splitting is about twice larger in Na<sub>2</sub>KSb (0.55 eV vs 0.34 eV in GaAs). These bands form initial states in the photoabsorption process.

Thus, the similarity of the band structures of the Na<sub>2</sub>KSb and GaAs allows us to conclude that the optical orientation of electron spins is possible in the Na<sub>2</sub>KSb, and, consequently, the photoemission of spin-polarized electrons.

In our previous work [38] we developed an image-type spin detector prototype for measuring the normal (to the surface of the detector) component of the electron beam polarization. This detector is based on the injection of spinpolarized free electrons from GaAs/(CsO) cathode into a heterostructure with  $GaAs/Al_xGa_{1-x}As$  quantum wells (QWs) and recording the circularly polarized cathodoluminescence (CL) with spatial resolution. A flat vacuum photodiode composed of two effective NEA semiconductor electrodes was designed and studied in Refs. [39-42]. In this Letter, we demonstrate that alkali antimonide photocathode can be utilized as an effective source of spinpolarized electrons and the anode—Al<sub>0.11</sub>Ga<sub>0.89</sub>As/Cs<sub>3</sub>Sb heterostructure, as the spin detector. Schematic presentation of the compact vacuum photodiode with a photo of the device and investigation of spin-dependent injection are shown in Fig. 2(a). The photocathode consists of an active polycrystalline Na<sub>2</sub>KSb layer of 140 nm thickness grown by vapor phase deposition on the glass of the input window and activated to NEA by Cs<sub>3</sub>Sb layer [43,44]. The semiconductor anode (target) comprises 200 nm Al<sub>0 11</sub>Ga<sub>0 89</sub>As layer with SiO coating, through which the structure is bonded to the glass of the output window. The final step of the cleaning procedure for the anode was carried out inside a glove box flooded with pure nitrogen, in which the anode was chemically treated in a solution of HCl in isopropanol [45]. The cleaned Al<sub>0.11</sub>Ga<sub>0.89</sub>As anode surface was also activated to the NEA state by coadsorption of cesium and antimony. Similar activation layer was utilized earlier for GaAs [46]. The photocathode and anode were planeparallel mounted in an air-tight manner on the opposite flat sides of a cylindrical alumina ceramics body. The diameters of the cathode and anode were 18 with a 1.0 mm gap between the electrodes. To compare spin-dependent



FIG. 2. Experimental setup. (a) Schematic drawing of optical setup for spatial CL polarization detection with a compact vacuum photodiode for the spin polarization measurements. Electrons emitted by  $\hbar\nu$ -photon excitation from the Na<sub>2</sub>KSb source are injected into an Al<sub>0.11</sub>Ga<sub>0.89</sub>As target. (b) Image of CL intensity distribution in the anode Al<sub>0.11</sub>Ga<sub>0.89</sub>As structure under injection of 2 eV electrons emitted from the photocathode at room temperature. (c) Cross-sectional view and a photograph of the assembled photodiode from the Na<sub>2</sub>KSb photocathode side, which can be used as a tablet-type source of spin-polarized electrons for accelerators. (d) Quantum yield as a function of the incident photon energy and (e) photoluminescence spectra from Na<sub>2</sub>KSb photocathode excited with the 1.59 eV (780 nm) laser diode. (f) Energy band diagram of both the Na<sub>2</sub>KSb source and the Al<sub>0.11</sub>Ga<sub>0.89</sub>As target with the NEA separated by a vacuum gap.  $E_c$  is the conduction band,  $E_v$  is the valence band and  $E_F$  is the Fermi level. (g) Circularly polarized ( $\sigma^+$ ,  $\sigma^-$ ) components of the PL spectra of Al<sub>0.11</sub>Ga<sub>0.89</sub>As target excited with the circular polarized light with 1.67 eV (740 nm) photon energy. (h) The corresponding circular polarization degree of the PL emission determined as  $P_{\rm PL} = (I_{\sigma^+} - I_{\sigma^-})/(I_{\sigma^+} + I_{\sigma^-})$ .

photoemission properties of Na<sub>2</sub>KSb/Cs<sub>3</sub>Sb photocathode with the GaAs one, we also constructed and studied the vacuum spin-photodiode with GaAs/(Cs, O) cathode and the same Al<sub>0.11</sub>Ga<sub>0.89</sub>As/(Cs, O) anode. Based on the developed vacuum diode [Fig. 2(c)], the tablet-type sources of spin-polarized electrons can be manufactured and used for accelerators. The evacuated photocathode source can be opened in the photoinjector, thus potentially eliminating the construction of photocathode growth chambers (Supplemental Material, Fig. S1 [47]).

Quantum yield (QY) of Na<sub>2</sub>KSb photocathode measured in the transmission mode as a function of the incident photon energy in combination with the photoluminescence spectrum are shown in Fig. 2(d). The QY is calculated as the ratio of the registered electrons per incident photons. The quantum yield has the threshold at about 1.4 eV and increases slightly in the range 1.4–2.5 eV reaching QY maximum at about 15%. The photoluminescence (PL) spectrum of the Na<sub>2</sub>KSb photocathode recorded at a room temperature [Fig. 2(e)] shows a peak with the energy of 1.42 eV (887 nm) which corresponds to the Na<sub>2</sub>KSb optical band gap transition in agreement with DFT calculation [Fig. 1(a)]. One can see that PL peak position coincides well with the photoemission threshold of QY spectra, meaning that the red edge in photoemission spectrum is determined by the energy gap rather than the work function of Na<sub>2</sub>KSb/Cs<sub>3</sub>Sb. Thus, the deposition of Cs<sub>3</sub>Sb layer on Na<sub>2</sub>KSb leads to formation of the NEA state as follows from QY, PL [Figs. 2(d) and 2(e)] and energy distribution curves measurements (Supplemental Material, Fig. S2). These results, together with earlier data on Cs<sub>3</sub>Sb photoemission properties [28], allow us to plot the band diagram shown in [Fig. 2(f)].

To characterize the Al<sub>0.11</sub>Ga<sub>0.89</sub>As/Cs<sub>3</sub>Sb target as a spin detector first we measured the circularly polarized PL and determined the spectral dependence of the degree of circular polarization of PL [Fig. 2(g)]. For *p*-Al<sub>0.11</sub>Ga<sub>0.89</sub>As layer of 200 nm thickness under excitation with photon energy  $\hbar \omega = 1.67$  eV the degree of PL polarization is about 22% that is close to the potentially feasible value of 25%. The results of polarized PL measurements show that *p*-Al<sub>0.11</sub>Ga<sub>0.89</sub>As can be used for spin polarimetry



FIG. 3. Circularly polarized ( $\sigma^+$ ,  $\sigma^-$ ) components of the PL spectra of Na<sub>2</sub>KSb photocathode excited with the circular polarized emission of laser diode with (a) 1.49 eV (780 nm) and (b) 3.05 eV (405 nm) excitations. The peaks at 1.42 eV in panels (a) and (b) correspond to the Na<sub>2</sub>KSb band gap transition. (c) and (d) The corresponding circular polarization degree of the PL emission determined as  $P_{\rm PL} = (I_{\sigma^+} - I_{\sigma^-})/(I_{\sigma^+} + I_{\sigma^-})$ . (e) The circular polarization degree of the PL emission of Na<sub>2</sub>KSb compared with GaAs (0.5  $\mu$ m thickness) as a function of photon excitation energy.

applications based on the optical detection of the freeelectron spin polarization similar to QWs structure used in Ref. [41].

In order to test the polarization properties of the Na<sub>2</sub>KSb/Cs<sub>3</sub>Sb photocathode, we measured circularly polarized spectra ( $\sigma^+$ ,  $\sigma^-$ ) of photoluminescence in the optical orientation mode. The polarized PL excited with the circular polarized emission of laser diodes with photon energy  $\hbar\omega = 1.49 \text{ eV}$  (850 nm) and  $\hbar\omega = 3.05 \text{ eV}$ (405 nm) is shown in Figs. 3(a) and 3(b), respectively. The corresponding spectral dependence of the circular polarization degree for the PL emission is shown in the bottom panels in Figs. 3(c) and 3(d). As can be seen, even at excitation energy of 3.05 eV the PL polarization is not zero (4%). Note that the broad unpolarized peak centered at 1.8 eV in Fig. 3(b) corresponds to the glass PL emission [51]. Dependence of the PL circular polarization degree on the excitation photon energy in Na<sub>2</sub>KSb in comparison with GaAs is shown in Fig. 3(e). At photon energy  $\hbar\omega =$ 1.49 eV the PL polarization showing a maximum, 23%, decreases rapidly with rising in the excitation energy up to ~2.4 eV. However, in contrast to GaAs, where PL polarization becomes zero, in Na2KSb it remains about 4% at higher photon excitation energies. Similarity in the polarization dependence of the PL at low excitation energies and in the band structures of Na<sub>2</sub>KSb and GaAs [Figs. 1(a) and 1(b)] suggests that the optical selection rules for these two semiconductors are the same (Supplemental Material, Fig. S3 [47]).

Thus, circularly polarized light can be used to create an unequal occupation of electrons in  $Na_2KSb$  with the spin orientation parallel and antiparallel to the propagation direction of the incident light. If the electrons escape from the multi-alkali photocathode  $Na_2KSb/Cs_3Sb$  into the



FIG. 4. (a) Circularly polarized ( $\sigma^+$ ,  $\sigma^-$ ) components of the CL spectra excited by the injection of spin polarized electrons emitted from the Na<sub>2</sub>KSb/Cs<sub>3</sub>Sb cathode at the accelerating voltage of 1.0 V. (b) The circular polarization degree of the CL emission determined as  $P_{\text{CL}} = (I_z^{\uparrow} - I_z^{\downarrow})/(I_z^{\uparrow} + I_z^{\downarrow})$ . (c) Image of the spin-integrated CL intensity, (d) difference between spin-up and spin-down CL intensities  $(I_z^{\uparrow} - I_z^{\downarrow})$ , and (e) 2D distribution of the derived CL circular polarization degree  $P_z$ . The images were taken at an accelerating voltage of 1 V. (f) Left axis: dependence of the CL circular polarization degree on the energy of injected spin-polarized electrons for the Na<sub>2</sub>KSb photocathode compared with the GaAs/(Cs, O) photocathode where a similar Al<sub>0.11</sub>Ga<sub>0.89</sub>As/(Cs, O) spin detector is used. Right axis: Dependence of the integrated nonpolarized CL (logarithmic scale) on the energy of injected electrons (green squares).

vacuum, as shown in the band diagram in Fig. 2(f), we can expect photoemission of spin-polarized electrons.

In order to test the Na<sub>2</sub>KSb/Cs<sub>3</sub>Sb photocathode as a source of spin-polarized electrons, the injection of photoemitted electrons into the Al<sub>0.11</sub>Ga<sub>0.89</sub>As/Cs<sub>3</sub>Sb heterostructure was studied using spin-polarized cathodoluminescence. Earlier it was shown that semiconductor heterostructures based on III-V compounds can be used as spin detectors of free electrons with spectral and spatial resolution [38,41,52–54]. The polarized CL spectra measured under injection of spin-polarized electrons with the energy of 1.0 eV are shown in Fig. 4(a). The maximum CL intensity corresponds to the radiation emission energy of 1.53 eV (810 nm), coinciding with the PL maximum of Al<sub>0.11</sub>Ga<sub>0.89</sub>As [Fig. 2(g)]. The degree of spectral circular polarization of the CL emission excited by the electrons with the energy of 1.0 eV is equal to 4.5% [Fig. 4(b)].

Recently we demonstrated that semiconductor spindetector allows to measure polarization with spatial resolution in direct image mode [38]. Figure 4(c) shows the image of spin-integrated CL intensity from two spots induced by two opposite spin-polarized electron beams injected into the  $Al_{0.11}Ga_{0.89}As/Cs_3Sb$  target [Figs. 2(a) and 2(b)]. The image is taken at an accelerating voltage of 1 V and reflects the electron distribution in the cross section. In this case, the CL polarization in the cross section of two electron beams should have the opposite sign and be determined by the polarization of photoemitted electrons exited by absorbed circularly polarized light in the photocathode. Figure 4(d) shows the CL intensity difference  $(I_z^{\uparrow} - I_z^{\downarrow})$ . Because of the injection of opposite polarized electron beams and its recombination in Al<sub>011</sub>Ga<sub>089</sub>As, the emitted CL contains both polarized emission components ( $\sigma^+$ ,  $\sigma^-$ ) with helicity oppositely distributed intensity in the two spots [Fig. 4(d)]: spin-up  $I_z^{\uparrow}$  and spin-down  $I_z^{\downarrow}$ . The CL polarization asymmetry image  $P_{\rm CL} = (I_z^{\uparrow} - I_z^{\downarrow})/(I_z^{\uparrow} + I_z^{\downarrow})$  is shown in Fig. 4(e). The detected  $P_{\rm CL}$  at electron injection with energy of 1 eV is 3% [Fig. 4(e)]. The dependences of the CL circular polarization degree determined from spectral and spatial distribution on the injected electron energy in the range of 0.6-4 eV are shown in Fig. 4(f).  $P_{CL}$  is maximal, of 9.5%, at low kinetic energies (0.6 eV) and monotonically decreases down to  $\approx 1\%$  as the electron energy increases to 4 eV. Comparing the CL polarization P<sub>CL</sub> for Na<sub>2</sub>KSb/Cs<sub>3</sub>Sb to GaAs photocathode [Fig. 4(f)] one can conclude that the latter is twice less. Assuming that the polarization of photoelectrons from GaAs cathode is in the range of 20%–25% [41], we can conclude that the polarization of photoemitted electrons from Na<sub>2</sub>KSb/Cs<sub>3</sub>Sb is in the range of 40%–50%.

The generation of spin-polarized electron beams with low emittance is essential for future accelerator applications [4,55]. We established that energy spreads of electron beams from Na<sub>2</sub>KSb/Cs<sub>3</sub>Sb cathode, generated by the light of energy lower than 1.3 eV, approach the limit imposed by the thermal energy of electrons and the quantum efficiency of the cathode is higher than that in metallic photocathodes (Supplemental Material, Fig. S4 [47]).

To summarize, the Na<sub>2</sub>KSb band structure was found to be very similar to GaAs with the same optical transition rules. However, the SOI gap in the VB of Na<sub>2</sub>KSb is almost twice larger than that in GaAs. Spin-polarized electrons in the Na<sub>2</sub>KSb photocathode were obtained by optical pumping with circularly polarized light and showed 23% polarization of photoluminescence and 40%-50% spin polarization of the emitted electrons. The  $Al_xGa_{1-x}As$ structure has also been shown to be promising as a spatially resolved spin detector for free electrons. High quantum efficiency and low emittance of spin-polarized electron beams provided by multi-alkali photocathodes make them of great interest for next generation high brightness photoinjectors. Finally, based on the created vacuum photodiode, the tablet-type sources of spin-polarized electrons can be manufactured and used for accelerators.

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