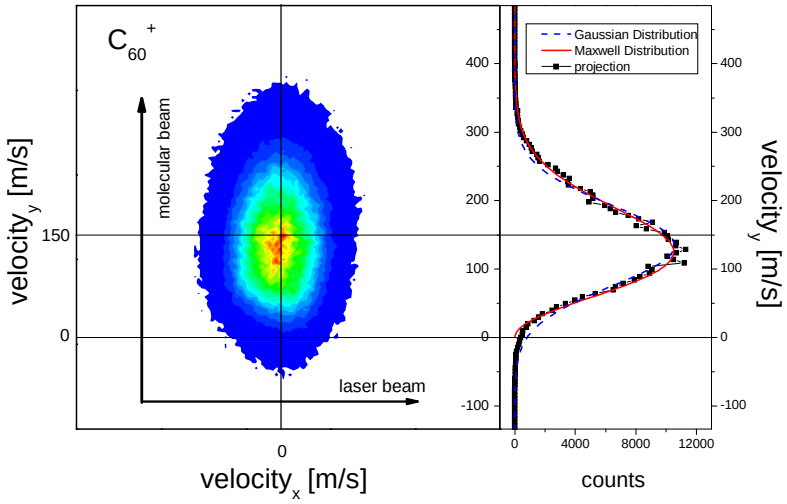

Figure 2.5: *Maxwell speed distribution of the C_{60} molecular beam. Left: spatial distribution of C_{60}^+ measured with Roentdek position sensitive detector. Right: projection of a thin slice of spatial distribution and Maxwell speed distribution fit. The best fit was with a temperature of 420° C, which is comparable to the measured oven temperature of 415° C.*

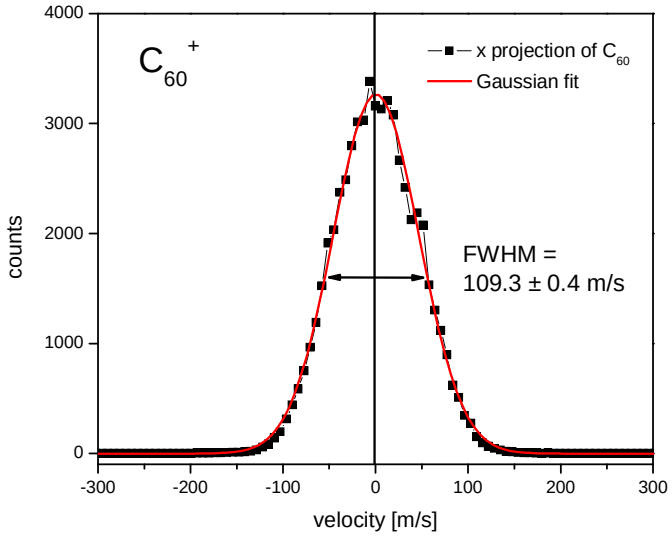

Figure 2.6: *A look at the x projection of the spatial distribution measured with a position sensitive detector. The projection is a thin slice of 5 pixels from the center of the distribution. The FWHM width of the Gaussian fit is 109.3 m/s, which corresponds to a length of 4.1 mm. Zero velocity is defined at the center of the Gaussian.*

Using an average velocity of 143 m/s found in Fig. 2.5, the lateral FWHM velocity corresponds to a beam divergence of $\sim \pm\frac{FWHM}{2*143} = 0.38$ rad or 22 degrees, which is larger than would be derived from the geometry indicated in Fig. 2.3.

An important factor to determine the width is the possibility of fragmentation after the acceleration. The kinetic energy release associated with the fragmentation process will increase the measured width. The interpretation of the widths will be further considered in Chapter 5.

The internal temperature has another important consequence for the interpretation of the results presented in this work. The C_{60} molecules iminating in thermal equilibrium, and considering the 3n - 6 = 174 vibrational degrees of freedom, the C_{60} molecule contains approximately 4.4 eV of internal energy before excitation with the femtosecond laser pulses. This can be calculated using the Eq. 2.5 [RWK01],

$$E_{in} = \frac{(3\widetilde{n} - 6)h\nu}{exp[h\nu/k_BT] - 1} \tag{2.5}$$

where $\widetilde{n}$ is the number of atoms in the fullerene and the average vibrational energy, ν, was taken to be that of the neutral system, i.e., 2.7x10^{13} Hz. The 4.4 eV is still small in comparison to the fragmentation threshold of C_{60} of 40 eV [HCF94].

2.2.2 Cold Molecular Beam Source

The internal temperature of C_{60} resulting from the effusive oven is 4.4 eV (see section about the oven). This energy is stored in vibrations which could mediate the absorption of energy. Therefore, a "Cold Source" apparatus, providing a molecular beam of C_{60} with a temperature of $\sim$ 80 K [HMH97, HHM96, HMB97], was brought to Berlin in a collaboration with the group of Prof. E.E.B. Campbell. An estimation of the population of vibrational levels at 80 K shows that nearly all molecules from the cold source remains in the ground vibrational level, with as little as 0.6 percent in the first excited state.

The source is similar to the cluster aggregation type sources used in the groups of Martin [ZMN94] and Haberland [ESS95]. The cold molecular beam is generated in an aggregation cell with liquid nitrogen cooled walls and filled with a helium buffer gas, as is shown in Fig. 2.7. A resistively heated oven sublimes C_{60} into the gas phase, which then has ample time for collisional cooling with helium, reducing the internal temperature.

The cold source was directly attached to the time-of-flight apparatus shown in Fig. 2.13, and the same extraction meshes and fields were used as for the linear or reflectron time-of-flight. Due to the high velocity of the C_{60} molecules in the cold molecular beams, deflection plates were introduced into the apparatus. These will be discussed in Section 2.5.

If the density of C_{60} is large enough, which is controlled by the oven temperature, clusters of C_{60} can form, as is seen in Fig. 2.8. The excitation source used in this case was a 100 fs, 800 nm Ti:Sapphire with a fluence of 1 J/cm^2. The broad peaks near masses of C_{120}^+ and C_{180}^+ indicate the formation of clusters of C_{60}. The substructure is a series of peaks separated by C_2 units. This fragmentation pattern may be explained by coalescence of the fullerene cluster [YHD93], followed by C_2 loss. If the temperature of the oven is low enough, i.e., at low density of molecules, clustering is no longer favorable, which ensures that only cold, isolated C_{60} is present in the interaction region. All photoelectron spectra taken with the cold source and presented in this thesis were taken in the regime where no clusters were formed, i.e., $\leq$ 470 °C.

2.3 Femtosecond Irradiation Sources

The primary sources of laser radiation used in this work to excite fullerenes are amplified Ti:Sapphire Laser systems. At the Max Born Institute, one has the capability of using various laser sources. The source is selected depending on the experimental question to be answered. The majority of results presented in this work used either the fundamental wavelength (800 nm) or the second harmonic (400 nm) generated in a second harmonic generation (SHG) crystal.

The basis of every system is a femtosecond Ti:sapphire oscillator. The output energy per pulse of these lasers are in the nano-Joule region, which is far too weak for the multiple photon excitation and ionization needed for C_{60}. Thus, the pulses need to be further amplified.

Each of these systems use chirped pulse amplification (CPA) to provide high power yet short pulse duration. CPAs circumvent nonlinear effects and damage thresholds by stretching the incoming, oscillator pulse in time and thus reducing intensity. After the pulse has been amplified, the pulse is temporally compressed.

The majority of these systems are regenerative amplification systems. Regenerative amplification seeds one of the stretched oscillator pulses into a cavity where it