Ultrafast electron relaxation dynamics in laser-ionized gases observed with time-resolved THz spectroscopy

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Abstract—**Ultrashort broadband THz pulses are applied to probe the electron dynamics of various gases following ionization by an intense femtosecond laser pulse. The dielectric function of the plasma is found to be Drude-like and yields the temporal evolution of the density and collision rate of the free electrons. The electron decay in a plasma with molecular ions such as O_2^+ is much faster than in monatomic plasmas like Ar^+/e^- due to dissociative recombination which is only possible in molecular plasmas. However, adding a small amount of the electron scavenger SF_6 to Ar substantially accelerates the electron decay and enables one to reliably determine the electronic temperature. Furthermore, anomalously high, metal-like electron collision rates are found. Kinetic plasma theory dramatically under-estimates these rates pointing towards additional velocity-randomizing processes like collective excitations.**

I. Introduction And Background

THE interaction of high-intensity ultrashort laser pulses and gaseous media is of great importance for a variety of fundamental phenomena and applications such as optical field ionization[1], the generation of high-harmonic radiation[2], and control of lightning in thunderstorms[3]. Detailed knowledge of the elementary processes and the charge-carrier dynamics is here of particular relevance. Examples are the mechanisms of the formation and loss of free electrons, and the plasma conductivity. In terms of a microscopic understanding, it is highly desirable to gain insights into the dynamics of plasma quantities like the density, temperature, and collision rate of the free electrons. Femtosecond (fs) time-resolved studies of gas plasmas employ an intense laser pulse to ionize a gas and subsequently probe the non-equilibrium conditions with a second pulse. Time-resolved THz spectroscopy has been proven as a powerful diagnostics method of laser plasmas[4,5]. In particular, the electron collision rate Γ and the plasma frequency Ω_{pl} of optically ionized gases lie in the THz range.

II. Experiment And Results

A typical experimental layout and the data evaluation performed in such measurements are described in detail elsewhere[4]. In brief, the plasma of interest is created by an 800-nm, 0.6-mJ, 20-fs pulse from an amplified Ti:sapphire laser system in a cell filled with either rare gases Ar, Kr or molecular O_2, N_2, H_2 or with SF_6-Ar mixtures and pure SF_6, respectively. The 100-fs THz probe pulse comprising frequencies from 10 to 25 THz crosses the transversally homogeneous plasma sheet and is detected amplitude- and phase-sensitively via electro-optic sampling.

The comparatively high probe frequencies allow us to extend the electron-density range accessible in the experiment up to $n_e \sim 10^{18}$ cm^{-3} since plasmas are only transparent above the plasma frequency. The measured plasma response is excellently described by both the Drude model and a micro-scopic approach based on the Boltzmann equation[4]. Fitting gives the temporal dynamics of the free-electron density n_e and the electron collision rate Γ. As shown in Figure 1, we observe a completely different relaxation behavior in the monatomic gas Ar compared to molecular O_2. The decay of the free-electron density in O_2 is substantially faster than in Ar. In addition, if only a small fraction of Ar is replaced by the electron scavenger SF_6, the electron lifetime in the Ar-SF_6 plasma is dramatically reduced. The reason for this observation is the lack of effective dissipation channels of the released energy of the ion-electron recombination in atomic systems as compared to molecular species with internal degrees of freedom such as vibrations or dissociation.

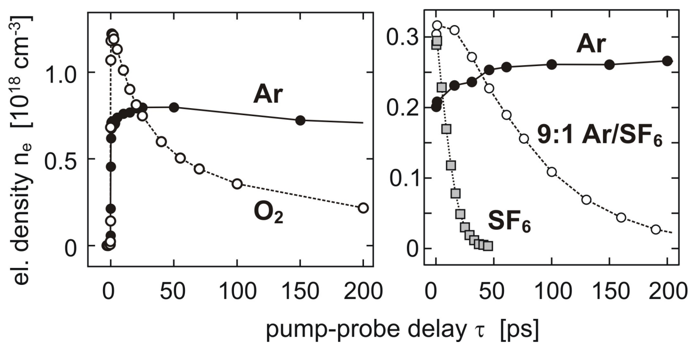

Fig. 1: Temporal evolution of the free-electron density n_e in various gases and gas mixtures after optical ionization. (left) Ar and O_2 provide comparison of the electron relaxation dynamics between atomic and molecular plasmas. (right) The admixture of SF_6 to Ar accelerates the electron decay considerably with time constants of 79 ps and 12 ps for (90%-Ar)/(10%-SF_6) and pure SF_6, respectively.

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