EVIDENCE OF LOW-BINDING-ENERGY ELECTRONS IN A LOW-TEMPERATURE HELIUM AFTERGLOW*

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(Received 12 July 1968)

Moderate, pulsed microwave heating of the free electrons in a weakly ionized helium afterglow plasma at 9.5 Torr at a gas temperature of 77°K produces an unusual modulation of the visible afterglow light. Observations of a second (probing) microwave signal indicate an accompanying significant increase in free-electron density under the assumption that there is no significant anomalous phase shift or absorption of the probing signal.

Observations have been made of the modulation of both visible afterglow radiation and microwave transmission resulting from perturbations of the free-electron gas temperature imposed during various intervals in the decay of a weakly ionized helium gas at a pressure of 9.5 Torr maintained at a gas temperature near 77°K. The perturbation of the free-electron temperature T_e is produced by the introduction of a second microwave signal of a different frequency and necessarily higher power than the probing microwave signal. This "heating" microwave signal is ≤ 200 μsec in duration. Under the tentative assumption that there is no significant anomalous phase shift or absorption of the transmitted probing microwave signal, interpretation of the modulation on phase shift and absorption implies a significant, unexpected increase in free-electron density n_e (on the order of unperturbed n_e) in the course of moderate microwave heating and a subsequent fast (~50-100 μsec) return to unperturbed n_e upon removal of heating.

The discharge tube is Pyrex, rectangular, approximately 24 cm in length, and makes a close fit within an X-band microwave waveguide. A 2-μsec, high-voltage (~7-kV) pulse applied between electrodes located outside the waveguide produces breakdown of the cataphoretically pure helium. At 200 μsec after breakdown, n_e is found to be $\sim 10^{11}$/cc, falling to 10^9/cc in approximately 4 msec. Spectrally resolved observations of visible light intensities in the early afterglow (within 800 μsec) were made with a Jarrell-Ash, Model 82-000, 0.5-m monochromator and an RCA phototube (S-11 photocathode). The decay of atomic He lines is markedly different than that of diatomic molecular He bands. The lines are very strong in the breakdown but decay rapidly in intensity. By approximately 300 μsec the sum of atomic line contributions to the signal from the photomultiplier can be neglected in comparison with the sum of molecular band contributions. The most prominent six molecular bands (5735 Å $[3\,^3\Delta_u\text{-}2\,^3\Pi_g]$, 4725 Å $[4\,^1\Sigma_u{}^+\text{-}2\,^1\Pi_g]$, 4650 Å $[3\,^3\Pi_g\text{-}2\,^3\Sigma_u{}^+]$, 4547 Å $[4\,^3\Sigma_u{}^+\text{-}2\,^3\Pi_g]$, ~4470 Å, and 4439 Å $[4\,^3\Pi_u\text{-}2\,^3\Pi_g]$)[1,2] can be followed with 15-Å resolution to beyond 800 μsec. These six bands appear to decay at the same rate. Beyond 800 μsec the observations of light intensity are not spectrally resolved; i.e., they are observations of the signal from the photomultiplier with total visible radiation incident on the photocathode.

When a short pulse of microwave radiation of insufficient power to cause significant ionization of ground-state or metastable-state He atoms is incident on an He afterglow plasma of gas temperature near 300°K at times on the order of a millisecond after the discharge, the visible afterglow light intensity is partially quenched during application of the pulse.[3,4] Generally such a pulse of constant power will maintain a constant free-electron temperature T_e (after the time necessary to establish this T_e) during its application even in the event that n_e decays appreciably during its application. If the pulse length is short (i.e., no significant change of n_e by recombination or diffusion losses occurs during application), then the afterglow light is observed to maintain a constant level until the removal of the microwaves. At this time the light intensity returns to approximately the same intensity as before introduction of the microwaves, in the time required for T_e to decay by means of elastic collisions between the free electrons and helium atoms.[4] The partial quenching of visible afterglow light intensity has been interpreted as due to a reduction of the recombination coefficient by increase in T_e which results in subsequent reduction of the light from recombination.

The upper traces of Fig. 1 illustrate the very different effect produced on the visible afterglow light of a 9.5-Torr, helium afterglow plasma at a gas temperature of 77°K. The electron temper-

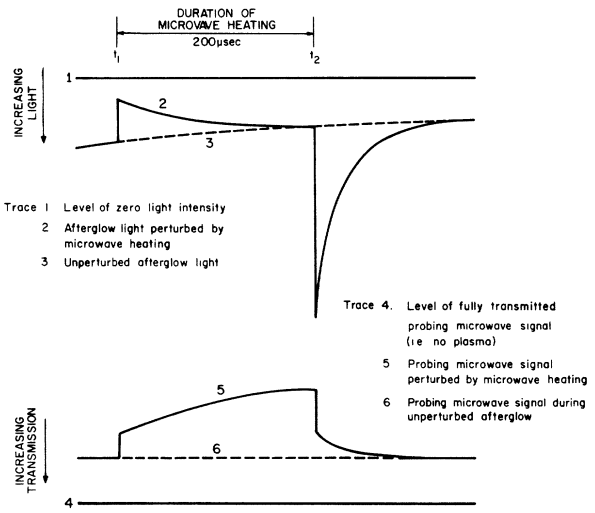

FIG. 1. Effects of moderate microwave heating upon the visible afterglow light (upper traces) and upon a transmitted probing microwave signal (lower traces) for a He afterglow plasma at a gas pressure of 9.5 Torr and gas temperature of 77°K.

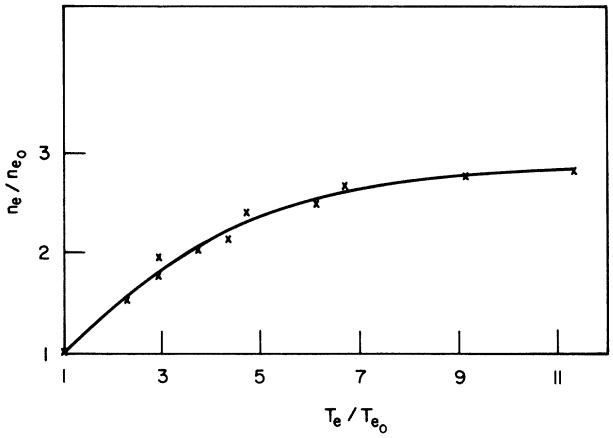

FIG. 2. Typical plot of electron density at 150 μsec of microwave heating versus electron temperature during heating. Each has been normalized to the respective unperturbed value for this particular afterglow time. ($T_{e_0} \sim 170$°K for this plot, gas temperature of 77°K and pressure of 9.5 Torr.)

ature increases rapidly (~3 μsec) at the time of application of the microwave pulse t_1, accompanied by an expected rapid partial quenching of the visible afterglow light. However, visible afterglow light subsequently increases and reaches a nearly constant amplitude within approximately 150 μsec. This amplitude is nearly coincident with the unperturbed amplitude when the heating is relatively small (this is the case in Fig. 1) and progressively less than the unperturbed amplitude as heating power is increased. At the time of removal of the heating microwaves, visible afterglow light intensity increases, within approximately 2 μsec, to an amplitude that can be several times the unperturbed and then decays to the unperturbed amplitude in approximately 100-200 μsec. Such a transient increase in afterglow light upon removal of heating microwaves was observed in He afterglows at gas temperatures of approximately 77 and 4.2°K by Goldan, Berlande, and Goldstein and termed the "afterpulse."[5]

The lower traces of Fig. 1 illustrate amplitude modulation produced on the low-power probing microwave signal (applied over a longer interval of time than the microwave heating pulse) due to application of the heating pulse. The rapid increase in the absorption at t_1 is in response to a rapid increase in T_e, inferred from the increase in the elastic electron-neutral collision frequency for momentum transfer as determined from absorption and phase-shift measurements on the transmitted probing microwave signal. The subsequent more gradual increase in absorption during application of the heating microwaves is found to result from an increase in $n_e - T_e$ remaining approximately constant. In the case of Fig. 1 the increase in T_e by heating is about 100% and the modulations of the probing microwave signal and the visible afterglow light are typical of this relative increase in T_e throughout the region where most observations were made, i.e., between 1 and 3 msec.

Figure 2 is a typical plot of n_e, as inferred from the absorption and phase-shift measurements, at 150 μsec of microwave heating versus T_e during heating, both normalized to their respective unperturbed values. It should be noted that T_{e_0} at 1 msec of the afterglow (i.e., unperturbed T_e at 1 msec) is still falling and well above 77°K (~190 and ~150°K at 2 msec).[6] Ambipolar diffusion enhanced by increasing T_e must be significant in determining the shape of the latter portions of the curve, but nonetheless n_e can be increased by a factor of almost 3 during microwave heating, and merely doubling T_e gives rise to an almost 50% increase in n_e.

When the heating microwaves are suddenly removed at t_2, T_e falls rapidly to a value near unperturbed T_e, accompanied by the almost discontinuous fall in absorption seen in Fig. 1. The more gradual change in absorption after this fall results mainly from a decrease in n_e to unperturbed n_e.

The modulation of the unresolved visible light intensity is assumed to involve no spectral redistribution of the observed radiation because the six major molecular bands listed earlier show apparently identical modulations, relative to their own unperturbed intensities, for early afterglow times where they can be spectrally resolved.

The preceding remarks were based on the assumption that the free-electron density and electron temperature measurements are reliable. However, the reliablility of determining free-electron density and electron temperature from phase-shift and absorption measurements on the probing microwave signal rests with the assumption that the phase shifts and absorptions are related to the free electrons undergoing elastic electron-atom collisions. The following are some additional possibilities:

(1) Inelastic electron collisions. However, at a given time in the afterglow a significant effect on phase shift or absorption due to free electrons undergoing inelastic collisions requires a population of "target" states several order of magnitude larger than the free-electron density or inelastic collision cross sections several orders of magnitude larger than the elastic electron–ground-state-atom collision cross section for momentum transfer.

(2) Resonant absorption of probing microwaves (i.e., absorption by means of photon-induced transitions between bound states). The probing microwave photon energy is small ($\sim 3\times 10^{-5}$ eV). Therefore it is necessary to consider states which are situated close in energy. It is difficult to explain the modulation of visible molecular afterglow light if states responsible for a resonant microwave absorption are solely states of the atom. Moreover, it appears that if the probing microwave signal is significantly absorbed as the result of probing microwave photons inducing transitions between molecular rotational levels, the density of molecules in such rotational levels would be nearly as large as the density of helium atoms initially present in the gas ($\sim 10^{18}$/cc).

(3) Particle redistribution or oscillation. The possibility that the observed perturbations are due to particle redistribution was examined in several ways: (a) by spatially resolved observations of afterglow light across the width and along the length of the discharge tube, (b) by heating in the TE_{20} microwave mode rather than TE_{10} and observing visible afterglow light, and (c) by observing visible afterglow light emitted by a discharge tube situated outside the microwave waveguide and irradiated by means of a microwave horn. There was no evidence in these cases to support the possibility of spatial redistribution.

Tentative acceptance of the reliability of the measurements of n_e and T_e leads to certain implications: The rise of afterglow light (after the initial partial quenching) in the course of heating could be explained on the basis of increasing rate of recombination, caused by an increasing n_e. The afterpulse of light could be understood as a response to the sudden increase in recombination coefficient, the result of a rapid decrease in T_e. It should be noted that since it is difficult to determine at what time n_e actually returns to unperturbed n_e, it should not be inferred from Fig. 1 that the light in the tail of the afterpulse continues above its unperturbed level beyond this time.

The functional dependence of n_e on time t after removal of the microwave heating before n_e returns to normal cannot be explained by invoking a simple recombination law. Assuming that $dn_e/dt \propto n_e^{\sigma}$, slopes of plots of $\ln(dn_e/dt)$ vs $\ln n_e$ give inconsistent and, at times, inordinately high values of σ ($\sim 3 < \sigma < \sim 5$), T_e being essentially constant. Moreover dn_e/dt within this period is much greater than can be expected on the basis of the decay rates of n_e observed in the unperturbed afterglow. The additional observations that n_e returns to unperturbed n_e and that dn_e/dt at the time of application of the heating microwaves t_1 increases monotonically with T_e (i.e., monotonically with microwave heating power) suggest the following interpretation: There is a population $[X]$ of weakly bound electrons in a near-collisional equilibrium with the free-electron gas at a given afterglow time. Microwave heating of the free-electron gas has the effect of disturbing this equilibrium by increasing the destruction rate of $[X]$ and perhaps reducing the rate of formation. The electron density increases during heating until a new equilibrium population of state or states X is established at the enhanced T_e maintained during heating. After the microwave heating pulse is removed, T_e falls to a value near unperturbed T_e and $[X]$ is subsequently re-established at the value previous to heating.

At present additional efforts are being considered to confirm further the existence of a significant population of weakly bound electrons.

*Work supported by National Aeronautics and Space

Administration under Contract No. NGR 14-005-037.

[1]G. Herzberg, Molecular Spectra and Molecular Structure: I. Spectra of Diatomic Molecules (D. Van Nostrand Company, Inc., Princeton, N.J., 1950), pp. 535-536.

[2]With the exception of 4470Å, the wavelengths given represent positions of the 0-0 bands. Herzberg's listing closest to the radiation observed to be maximum in the vicinity of 4470Å is a 0-0 band at 4456 Å [$4^3\Sigma_u^+ - 2^3\Pi_g$].

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[5]P. D. Goldan, J. A. Berlande, and L. Goldstein, Phys. Rev. Letters 13, 182 (1964).

[6]Assuming an elastic electron-neutral collision cross section for momentum transfer of 5.3×10^{-16} cm^2 from J. L. Pack and A. V. Phelps, Phys. Rev. 121, 798 (1961).

THEORY OF NUCLEATION RATES*

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(Received 15 July 1968)

A general procedure is outlined for the first-principles calculation of the rate of decay of a metastable phase.

One of the more important outstanding problems in statistical mechanics involves the rate of decay of a metastable phase. It would be very useful to know how to make a first-principles calculation of, for example, the rate of condensation of supersaturated vapor, the realignment of a magnetic domain in an applied field, or the decay of a persistent current in a superfluid. All of these processes occur when the system undergoes a statistical fluctuation large enough to nucleate the phase transition. The characteristic fluctuations for the three transitions mentioned above are, respectively, a liquid droplet, a cluster of reversed spins, and a vortex ring.

The problem is an old one. Probably the most important piece of work in the field is the calculation by Becker and Döring in 1935 of the condensation rate for a supersaturated vapor.[1,2] In the absence of a first-principles derivation, however, this semiphenomenological calculation has been subject to frequent criticism, some modern authors claiming correction factors of order 10^{15} or more.[3] Interest in the nucleation theory has been further stimulated recently by its apparently successful application in understanding the onset of resistivity in superfluids[4] and superconductors.[5] Here again, existing theories are phenomenological in certain important aspects, and contain completely unknown factors.

In the following note, we outline a scheme for the calculation of nucleation rates for a class of simple but nontrivial models of phase transitions. The basic ideas seem to be quite general. Work is now in progress on the practical application of this scheme to the calculation of the rate of current-reducing fluctuations in a superfluid.

We consider a system described by a set of classical variables $\eta_i, i = 1, 2, \cdots N$, where N is the number of degrees of freedom. For example, η_i could be the magnetization at the ith site of a magnetic lattice, or the order parameter at the ith position in a superfluid. For the sake of simplicity, we assume that the η's are real numbers varying from $-\infty$ to $+\infty$. It will greatly simplify the following analysis if we further assume that, like an Ising or spherical model, the system under consideration has no internal dynamics of its own. That is, in the absence of interactions with a heat bath, the configuration $\{\eta\}$ remains fixed. It turns out to be fairly easy to generalize this calculation to cases where the motion of $\{\eta\}$ is governed by, say, Newton's laws or the Ginzburg-Landau equation. But such motion is not of direct interest here because it is always energy conserving, whereas first-order phase transitions are nucleated by fluctuations which do not conserve energy.

Next, we assume that, when in interaction with a constant-temperature bath, the system will make a transition from $\{\eta'\}$ to $\{\eta\}$ with a probability P per unit time of the form

$$P(\{\eta\},\{\eta'\})dt = \Gamma\left(\frac{dt}{\Delta}\right)(\pi\Delta)^{-\frac{1}{2}N}\exp\left(-\frac{1}{2kT}E\{\eta\}\right)\exp\left(\frac{1}{2kT}E\{\eta'\}\right)\exp\left[-\frac{1}{\Delta}\sum_i(\eta_i-\eta_i')^2\right], \quad (1)$$