Potential electron emission induced by multiply charged ions in thin film tunnel junctions

D. A. Kovacs

Experimentalphysik II, Ruhr-Universität Bochum, 44801 Bochum, Germany

T. Peters, C. Haake, M. Schleberger, and A. Wucher Fachbereich Physik, Universität Duisburg-Essen, 47048 Duisburg, Germany

A. Golczewski and F. Aumayr

Institut für Allgemeine Physik, Technische Universität Wien, A-1040 Vienna, Austria

D. Diesing*

Physikalische Chemie, Universität Duisburg-Essen, 45117 Essen, Germany (Received 3 March 2008; revised manuscript received 7 May 2008; published 23 June 2008)

Thin film metal-insulator-metal tunnel junctions are used to investigate the electronic excitation process induced by the impact of multiply charged ions onto a metallic surface. Hot charge carriers (electrons and holes) generated by the dissipation of the kinetic and potential energies of the projectiles are detected as an ion induced internal emission current from the bombarded "top" metal film into the "bottom" substrate electrode. Results are presented for Ar^{q+} ions with a kinetic impact energy of 1 keV and charge states q=1-8 impinging onto an Ag-AlO_x-Al junction. It is shown that the internal emission current, whereas this influence between the two metal films is found to strongly influence the internal emission current, whereas this influence becomes much weaker with increasing projectile charge state. The results are shown to be qualitatively well described in the framework of a thermodynamical free-electron model.

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I. INTRODUCTION

The interaction of charged particles with solid surfaces is an intensively investigated field. The underlying physical processes are of great interest not only for basic research, but also for technological applications implying material modifications such as sputtering, melting, evaporation, etc. During the interaction, ions loose energy due to nuclear and electronic stopping,¹ de-excitation via Auger transitions,² plasmon excitation,³ and photon emission.⁴ At low kinetic energies, e.g., $E_{\rm kin} < 200$ keV for Ar in Ag, nuclear stopping dominates over electronic stopping.⁵ Nevertheless, electronic excitation is an important dissipation channel due to Auger de-excitation, especially in the case of multiply charged ions (MCI).⁶

Intensive research on the interaction of multiply charged ions with metallic surfaces (see Refs. 7 and 8 and the references therein) has lead to a detailed understanding of the associated processes. In addition to the kinetic energy which is introduced into the solid upon impact, MCI carry potential energy which is stored in form of its ionization energy. The MCI can therefore be used to carry a huge amount of energy into a very small volume. This may give rise to various interesting phenomena such as nonlinear processes or surface modifications. In order to study the effects induced by the potential energy of the projectile, the kinetic impact energy has to be sufficiently low. The collision of the MCI with the solid can then be described as a four stage process; three of them occurring above or close to the surface and the fourth below the surface. The first three stages include the image charge acceleration of the projectile toward the surface⁹ and the formation of a so-called hollow atom:^{10,11} below a certain

distance, multielectron capture into highly excited Rydberg states of the projectile occurs by resonant electron transfer from the conduction band of the solid. This results in a dynamically neutral projectile with empty inner shells. Ongoing electronic transitions, such as resonant neutralization and autoionization, together with level shifting due to screening and image interaction, lead to a projectile highly transient in nature. Up to here, mainly slow electrons are emitted from the projectile as well as from the surface. Closer to the surface, the filling of the inner shells starts leading to the emission of high energy Auger electrons. Upon surface penetration, the electrons in outer shells of the projectile are peeled off and the projectile is effectively screened by the metal electrons.^{12,13} At this stage, a secondary hollow atom forms inside the solid. Since the time spent in front of the surface is not sufficient for a complete relaxation, it still possesses empty inner-shell states which are now filled by Auger neutralization, or in the case of heavier elements, by competitive x-ray emission. Depending on the experimental details such as projectile type, initial charge state, and angle of incidence, the subsequent de-excitation of the hollow atom below the surface results in the emission of Auger electrons with varying energies. The last step in the de-excitation will be the filling of the K shell associated with the emission of KLL-Auger electrons. At lower values of the charge state q, i.e., for Ar^{q+} with $q \leq 8$, an apparently linear dependence of the number of electrons emitted into the vacuum per impinging ion (external emission yield) on the potential energy of the MCI has been found.^{14–16} For q > 8, the yield showed rather a linear dependence on the charge state q.¹⁴

By means of metal-insulator-metal (MIM) tunnel junctions, hot electrons produced in the exposed (top) metal film can be detected in the opposite (bottom) metal as an internal emission current, if they succeed to overcome the potential barrier provided by a very thin oxide layer separating the two metals.¹⁷ Thus, the detection occurs very closely to the excitation region and, therefore, all electrons overcoming the potential barrier can be detected. Unlike the external emission, also hot holes may be detected in such a MIM device, which give a negative contribution to the internal emission current. The detection sensitivity of electrons may be increased or decreased relative to that of holes by applying a bias voltage between the metal films and, hence, by varying the average barrier height for electrons and holes.¹⁷ The resulting ion induced current-voltage dependence reveals information on the energy distribution of the hot charge carriers in the topmetal film. The internal emission current also depends on the potential barrier parameters, i.e., barrier width and thickness. These can be modified by changing the oxidation procedure during preparation, by varying the parameters of the oxidation, or by selecting another metal oxide to act as an insulator layer. Recently, we found that the net number of electrons emitted per impinging ion from the top into the bottom-metal layer (internal emission yield) depends also approximately linearly on the potential energy of the MCI.¹⁸

The aim of the present work is to investigate the internal electron emission, induced by slow multiply charged ions impinging onto the top surface of a MIM junction. For this purpose, we employed an Ag-AlO_x-Al junction, which was successfully applied in our groups to investigate the internal emission of charge carriers induced by photons, singly charged ions, and neutrals.^{17,19,20} We present experimental results on the internal emission yield, in particular, its dependence on potential energy and bias voltage, respectively, and discuss them in the framework of a thermodynamical free-electron model.

II. EXPERIMENT

The MIM junctions employed here consist of a 50 nm aluminum film evaporated onto an insulating glass substrate, an insulating layer of about 4 nm thickness formed by local oxidation of the aluminum film, and a 20 nm silver film on top (see Fig. 1). Details of how these devices are produced and characterized have been published elsewhere.²¹ From the measured capacity of the junctions, $C \approx 70$ nF, the relative dielectric permittivity of the oxide layer was estimated to $\epsilon_r \approx 8$.

All experiments were performed under high-vacuum conditions (typically $p=5 \times 10^{-8}$ mbar). The samples were irradiated under normal incidence with multiply charged Ar^{*q*+} ions produced by an ECR ion source of 14.5 GHz described in detail elsewhere.²² The ion beam was pulsed with a typical pulse width of 1.5 s. By changing the extraction potential of the ECR source, the samples were bombarded with ions of constant kinetic energy while varying the charge state. Under the applied conditions, no changes of the current-voltage characteristics of the tunnel junction have been observed during the experiments.

During irradiation of the top Ag film of the Ag-AlO_x-Al junctions, the ion induced current I_T , flowing from the Ag to



FIG. 1. (Color online) Schematic drawing of the Ag-AlO_x-Al thin film tunnel junction, showing the silver face being bombarded by multiply charged argon ions.

the Al layer, was measured directly, but was recorded as a voltage signal by using a current-to-voltage converter with a conversion factor of 10 mV/pA. This current is given by the emission of hot charge carriers (electrons and holes) across the barrier, being the sum of the (positive) electron and the (negative) hole emission currents I_e and I_h , respectively. Between the two metals, a bias voltage U may be applied, such that for U > 0 (U < 0), the Ag film is at a negative (positive) potential with respect to the aluminum film. By applying a bias voltage, a bias induced electron current Ibias was measured. Up to ± 0.3 V, I_{bias} remained below the detection limit of ± 5 pA. By working in pulse mode, I_T could be determined, at higher bias voltages, by subtracting I_{bias} from the total current measured in the bottom Al film. The bias voltage was limited to the range between -1.5 and 1.5 V in order to avoid a damage of the insulating layer.

The *internal electron emission yield* γ is introduced to characterize the efficiency of ion induced electron emission in MIM junctions, being defined as the net number of negative elementary charges per impinging ion flowing across the oxide barrier. It can be determined as

$$\gamma = I_T / (I_q / q), \tag{1}$$

where q is the ion charge state (selected by a bending sector magnet) and I_q is the primary ion-beam current (measured by a Faraday cup). Although the carrier emission is only partially due to quantum-mechanical tunneling, we will use, for simplicity, the term "tunneling yield (current)" instead of "internal electron emission yield (current)." Since the detected hot charge carriers are generated due to kinetic and potential-energy dissipations in the top-metal layer, the tunneling yield can be defined as the sum of a kinetic and a potential tunneling yields γ_k and γ_n , respectively.

III. RESULTS

In Fig. 2 the tunneling yield γ , induced by Ar^{*q*+} multiply charged ions with a kinetic energy of 1 keV is plotted against the potential energy E_{pot} of the projectile. The yield increases monotonously with increasing charge state and can be well fitted by a linear function. The observed linear dependence is



FIG. 2. Tunneling yield γ , induced by Ar^{*q*+} multiply charged ions in Ag-AlO_{*x*}-Al, plotted against the total potential energy E_{pot} . The solid line is a linear fit to the experimental data (squares). The kinetic energy of the ions was 1 keV.

in good agreement with previous results on external electron emission, measured for comparable MCI charge states and velocities.^{14–16}

From the straight-line fit indicated in Fig. 2, an energy specific potential yield $\gamma_p/E_{\rm pot} \approx 10^{-3}/{\rm eV}$ can be derived. This can be viewed as the efficiency of the potential energy to produce a tunneling current, which we have shown to be practically independent of the impact velocity of the MCI up to kinetic energies of 12 keV.¹⁸ A comparison with the values of the external electron emission yield γ_{ext} obtained on other metal surfaces, such as W,¹⁵ Mo,¹⁶ and Au,¹⁴ shows that the efficiency for internal emission in our MIM junction is by about 1 order of magnitude smaller than that for external emission. One reason for the lower emission yield could be the fact that beside electrons, also holes, which give a negative contribution to the tunneling current, are excited in the metal film. It was shown, however, that in Ag-AlO_y-Al tunnel junctions, the average potential barrier for holes is by about 1 eV higher than for electrons.^{23,24} This suggests the contribution of holes to the tunneling current to be relatively small when no bias voltage is applied between the metal films and, hence, it cannot explain the large difference. A more plausible explanation may be related to the transport of the hot charge carriers. Since argon ions with a kinetic impact energy of 1 keV may penetrate the silver film only up to a depth of at most 6–7 nm,⁵ hot charge carriers produced in the Ag top layer have to travel over longer distances in order to reach the bottom Al layer than in order to reach the vacuum. Hereby, electron-electron scattering may act to attenuate the detected electron current, as was clearly shown in experiments on internal electron emission induced by photons in similarly prepared Ag-AlO_x-Al samples.¹⁷

The kinetic electron emission yield γ_k can be estimated by extrapolating the straight-line fit to $E_{\text{pot}}=0$ in Fig. 2. The resulting $\gamma_k \approx 10^{-2}$ or $\gamma_k/E_{\text{kin}} \approx 10^{-5}$ eV⁻¹ can be safely neglected when compared with the potential electron emission yield γ_p . This result is not very surprising, since several factors may contribute to this lack of efficiency. First, while the potential energy of the projectile is directly transferred via Auger processes to individual electrons in the solid, the ki-



FIG. 3. Bias dependence of the normalized tunneling yield, induced by Ar^{q+} in a Ag-AlO_x-Al junction, for selected charge states q. The kinetic energy of the ions was 1 keV. The connecting lines are drawn to guide the eye.

netic energy is transferred gradually, during solid penetration, to several electrons from the surrounding region. In the latter case, the energy is distributed, along the trajectory of the projectile, among a larger number of electrons, but having lower kinetic energies. As a consequence, most of the excited electrons might not have enough energy to overcome the potential barrier. A more detailed investigation on kinetically induced electronic excitation in tunnel junctions will be presented in a forthcoming paper. Another reason for the reduced efficiency is that an important part of the kinetic energy of the projectile is lost in collisions with target atoms.

Figure 3 depicts the dependence of the normalized tunneling yield $\gamma(U)/\gamma(0 \text{ V})$, induced by Ar^{*q*+}, on the bias voltage U, for selected values of the charge state q, measured at a constant kinetic impact energy of 1 keV. A strong dependence of the bias voltage characteristics on the charge state can be observed. For q=1, the tunneling yield changes sign at $U_0 = -1.05$ V. We already found such a sign reversal in the irradiation of a similarly prepared Ag-AlO_x-Al junction by Ar⁺ ions with kinetic energies of 12 keV,¹⁷ as well as in photo conduction experiments.¹⁷ We interpret this sign reversal as follows: At relatively low energies dissipated into the electronic system, the energy distribution functions for electrons and holes are symmetrical relative to E_F . If the barrier heights Φ_e and Φ_h for electron and hole emissions, respectively, were equal, the hole emission would counterbalance the electron emission at $U_0 \approx 0$ V. The approximate sign is due to the slight distortion of the symmetry caused by the monotonous energy dependence of the density of electronic states. In the case $\Phi_e \neq \Phi_h$, a bias voltage $U_0 \approx (\Phi_e - \Phi_h)/e$ has to be applied in order to equalize the two barrier heights and, hence, to achieve a sign reversal of the tunneling current.

At higher charge states, no sign reversal of the tunneling yield is observed in Fig. 3 in the investigated voltage range; the normalized yield showing a weaker dependence on the voltage with increasing q until it becomes practically constant within the experimental errors at q=8. In this case, the increased potential energy causes a broader excitation spec-

trum of electrons and holes. While there is no upper limit for the excitation energy of electrons, the energy of an excited hole is limited by the finite width of the conduction band. In the free-electron-gas picture, the maximum possible hole excitation energy amounts to the Fermi energy (5.5 eV for Ag).²⁵ As a result, the electron-hole energy distribution becomes asymmetrical. It may be argued that, in a real metal, holes can be excited in core levels below the bottom of the conduction band. These holes, however, cannot move freely in the metal and have a finite lifetime, leading to the creation of secondary electron-hole pairs and, hence, further contributing to the asymmetry of the electron-hole pair energy distribution. As a consequence, hole emission becomes less effective to counterbalance the electron emission at equal barrier heights, and a higher bias voltage U_0 is necessary to zero the measured tunneling current.

IV. DISCUSSION

A. Electron temperature

The apparently linear relationship between γ and $E_{\rm pot}$ (see Fig. 2) is not a characteristic of the MCI-MIM interaction, since a similar behavior was found for $\gamma_{\rm ext}$ on metal surfaces, too.^{15,16} Qualitatively, this linearity was explained as follows: The de-excitation of the MCI occurs by a large number of Auger transitions, each one leading to the emission of electrons with energies in the range 15–30 eV. Thereby, the number of emitted electrons is determined by the number of Auger transitions, which is proportional to the potential energy. We will show below that this linear behavior may be understood also within a simple thermodynamical model.

In the present paper, an attempt is made to give a more quantitative explanation of the $\gamma(E_{\rm pot})$ dependence obtained in both internal and external electron emission experiments. The reason why the two kinds of experiments can be treated likewise is that the electron emission is more or less isotropic and that the oxide layer of the MIM introduces a barrier for the electrons in a similar way as the metal surface. We have shown recently¹⁷ that tunneling processes play only a minor role in the internal electron emission, which is, hence, dominated by "over-the-barrier" processes. The theoretical approach chosen here was inspired by the thermionic emission, where electrons are ejected into the vacuum from metal surfaces heated to several thousand Kelvin.²⁶

In the present model, we assume that the MCI impact causes a local heating of a small volume δV of the electronic system and that the resulting electron energy distribution in δV can be approximated by a Fermi distribution. To first order, we assume δV to be independent of the charge state of the MCI. Even though higher projectile charge states will lead to the excitation of larger numbers of "primary" Auger electrons, the present model assumes a fast redistribution, on a time scale of a few femtoseconds, of the dissipated energy among the electrons within δV , and therefore the entire energy deposition is parametrized by the transient local electron "temperature" within this volume. Moreover, transport of the generated hot carriers within the top Ag film is neglected in the present level of approximation, and therefore the (internal and external) electron emission is treated as if the "hot spot" was located directly at the respective interface.

Under the conditions defined above, the excited electrons are described by the Fermi-Dirac distribution function

$$f_{\rm FD}(E) = \frac{1}{e^{\beta(E-\mu)} + 1},$$
 (2)

where E is the electron energy relative to the bottom of the conduction band, μ is the chemical potential of the hotelectron gas, being equal at low temperatures with the Fermi energy E_F , and β is a slope parameter. Already, Hagstrum² suggested the local heating of the electronic system by impinging noble gas ions as a possible electron ejection mechanism. Šroubek²⁷ introduced an electronic temperature to describe the electron emission also from semiconductor surfaces as GaAs. Later, Šroubek et al.²⁸ used the same term within a thermal hot-spot model and estimated an electronic temperature of 14 000 K when a copper surface is bombarded by 1 keV Na⁺ ions. Recently, we have shown that electron temperatures as high as 12 000 K are required to explain the internal emission in MIM junctions.¹⁷ From a thermodynamic point of view, it is, however, questionable if such an electronic temperature may be introduced on the short time and length scales (femtoseconds and nanometers, respectively) of relevance here, since only a few electrons are involved in the excitation process. Therefore, Lindenblatt et al. avoided this term and used instead the term "slope parameter" or "fictitious temperature" in order to describe the electron energy distribution function at low excitation energies.²⁹ They calculated a value of 3000 K for projectile kinetic energies of 10 eV.

Independently on the used terminology, it was shown both theoretically²⁹ and experimentally²⁸ that the electron energy distribution spectra can be well fitted by the relation

$$n(E') \propto e^{-\beta E'},\tag{3}$$

where E' is the electron energy relative to the Fermi level of the metal. In order to enable the description of the equilibrium energy distribution function of the excited charge carriers, we introduce, with the required reservations arising from the above discussion, an "electronic temperature" T_e , given by the relation

$$\beta = 1/k_B T_e, \tag{4}$$

where k_B is the Boltzmann constant.

We used relation (3) to estimate T_e in the case of external electron emission induced by multiply charged ions at metal surfaces. For this purpose, we made use of the results of Kost *et al.*³⁰ who recently investigated the potential-energy dissipation during multiply charged Ar^{q+} -ion bombardment of copper. We replotted their Fig. 1 such that on the ordinate axis, the electron energy distribution function n(E)=dN/dE is plotted instead of EdN/dE and extracted T_e from fits to the high energy tails. As a result, we find electronic temperatures of 35 000 K (for q=2) up to 160 000 K (for q=8). These are huge values, being comparable with the Fermi temperature T_F of a free-electron gas, which, for Ag, is about 64 000 K.²⁵ Figure 4 depicts the relation between the electron temperature deduced from Ref. 30 and the total potential



FIG. 4. Temperature of the electronic excitation induced by bombardment with Ar^{q+} of 720 eV, plotted against the potential energy of the Ar^{q+} ions. The data (squares) are derived from Fig. 1 of Ref. 30. The solid line represents a linear fit to the data.

energy E_{pot} of the argon ions. A linear dependence of T_e on E_{pot} is apparent. The linear fit in Fig. 4 yields $T_e(E_{\text{pot}}=0) = 2.7 \times 10^4$ K and $\partial T_e / \partial E_{\text{pot}} = 223$ K/eV. The former value may be interpreted as being the temperature induced by the kinetic impact energy of the projectile, while the latter gives the temperature increase due to increasing potential energy.

B. Chemical potential

For a qualitative understanding of this linear dependence, we assume the electronic excitation to be represented by a free-electron gas described by the Fermi-Dirac distribution function given in Eq. (2). At the high electron temperatures estimated from Ref. 30, the chemical potential μ in the Fermi-Dirac function (2) cannot be approximated anymore by E_F , but becomes strongly temperature dependent. While up to about 20 000 K, μ is still well approximated by³¹

$$\mu(T_e) = E_F \left[1 - \frac{\pi^2}{12} \left(\frac{T_e}{T_F} \right)^2 \right],\tag{5}$$

where at higher temperatures, it can be determined only numerically by considering the conservation of the electron number density n_e , which is defined as

$$n_e = \int \frac{1}{4\pi^3} f[E(\mathbf{k})] d\mathbf{k}, \qquad (6)$$

where $f[E(\mathbf{k})]$ is the Fermi-Dirac distribution function defined in Eq. (2) and $d\mathbf{k} = dk_x dk_y dk_z$. The numerical solutions $\mu(T_e)$ calculated from Eq. (6) are plotted in Fig. 5(a), being compared with the values obtained in the case of a Maxwell-Boltzmann distribution. It can be seen that μ decreases monotonously with increasing temperature and changes sign at a value of the temperature close to T_F . While in the temperature region between 0 and T_F , μ still looks squarelike; at higher temperatures, it deviates from this behavior, approaching the expression derived in the limit of a nondegenerate electron gas



FIG. 5. Temperature dependences of the (a) chemical potential, (b) internal energy per unit volume, and (c) specific heat per unit volume of a Fermi-Dirac (connected stars) and a Maxwell-Boltzmann electron gases (solid line), respectively.

$$\mu(T_e) \approx k_B T_e \ln\left(\frac{1}{2}n_e \lambda_T^3\right) \Longrightarrow e^{\beta\mu} \propto T_e^{-3/2} \to 0, \qquad (7)$$

where $\lambda_T = h / \sqrt{2 \pi m k_B T_e}$ is the thermal wavelength.

C. Internal energy

Since the main dissipation channel for the potential energy is given by Auger de-excitation processes, it is clear that the potential energy is initially dissipated in the electronic system of the metal before it is transferred to the lattice via electron-phonon coupling. In Ref. 30, it has been shown that a fraction of about $(80 \pm 10)\%$ of the potential energy of an Ar^{q+} ion $(q \leq 9)$ is deposited in the bulk of copper. This suggests that the ion mainly de-excites upon penetration of the solid.

The temperature increase due to the energy dissipation leads to an increase of the internal energy of the Fermi gas. The internal energy u per unit volume of the electron gas is defined as

$$u = \int \frac{1}{4\pi^3} E(\mathbf{k}) f[E(\mathbf{k})] d\mathbf{k}.$$
 (8)

In Fig. 5(b), the internal energy per unit volume $u_{\rm FD}$ calculated for an ideal Fermi-Dirac (FD) electron gas is compared with the internal energy per unit volume $u_{\rm MB}$ calculated for an ideal Maxwell-Boltzmann (MB) distribution of electrons; both gases being described by an electron number density $n_e = 5.86 \times 10^{28} \text{ m}^{-3}$ (corresponding to Ag).³² It can be noticed that, at low temperatures, $u_{\rm FD}$ strongly differs from $u_{\rm MB}$, while, at high temperatures, $u_{\rm FD}$ approaches asymptotically $u_{\rm MB}$ due to the fact that the Fermi gas becomes gradu-

ally nondegenerated. Another observation is that between 4 and 16×10^4 K, $u_{\rm FD}$ can be well approximated by a linear function. Such a linear fit gives a slope value of 7.25 $\times 10^{24}$ eV/m³ K which is by about 5% smaller than the slope of the $u_{\rm MB}(T)$ curve. The validity of the linear approximation can be checked by plotting the specific heat per unit volume $c_v = du/dT_e$ as a function of temperature as shown in Fig. 5(c). It varies only by 15% in the temperature range $4-16 \times 10^4$ K.

Assuming that the potential energy is entirely dissipated into the electronic system, the increase of the internal energy per unit volume due to E_{pot} can be written as

$$\delta u(T_e) = u(T_e) - u(T_e^0) = E_{\text{pot}}/\delta V, \qquad (9)$$

where T_e^0 is the temperature of the excited Fermi gas at $E_{\text{pot}}=0$, being dependent on E_{kin} , and δV is the electron volume excited in the time interval δt . From Eq. (9), one can derive

$$\frac{1}{\delta V} = c_v(T_e) \frac{\partial T_e}{\partial E_{\text{pot}}} \bigg|_{E_{\text{kin}} = \text{const.}}$$
(10)

One may observe that, at high electron temperatures, where $c_v \approx \text{const.}$, Eq. (10) holds if $\partial T_e / \partial E_{\text{pot}} \approx \text{const.}$ This would actually confirm the experimental results shown in Fig. 4, indicating that $\delta V = \text{const.}$ is an appropriate assumption of the model. Equation (10) also offers an estimative value for δV . For copper, the derivative can be determined from the linear fit in Fig. 4 to 223 K/eV, while the specific heat per unit volume is about $1.0 \times 10^{25} \text{ eV/m}^3$ K, resulting in $\delta V = 448 \text{ Å}^3$. This corresponds to a radius of 4.75 Å and to a number of excited electrons per ion $N_{\text{exc}} = n_e \delta V \approx 38$.

D. Internal electron emission yield

In the following, we will describe the electron emission through the oxide of a MIM junction by assuming a local heating of a free-electron gas in the top-metal film, from room temperature to a temperature T_e . Further, we assume the excitation to be in thermal equilibrium for a time δt and the emission process to be stationary during this time. Under this conditions we may define a tunneling current density jwhich is flowing from the top to the bottom-metal electrode of a MIM junction as the sum of (positive) electron and (negative) hole current densities j_e and j_h , respectively. The former quantity can be written as (see Appendix A)

$$j_e = \frac{em}{2\pi^2\hbar^3} (k_B T_e)^2 \sum_{n=1}^{\infty} (-1)^{n-1} \frac{1}{n^2} e^{-n\Phi'_e/k_B T_e}, \qquad (11)$$

where *e* is the elementary charge, *m* is the effective mass of the electron (taken here to be equal to the free-electron mass), and Φ'_e is defined as

$$\Phi'_e = \Phi_e + E_F - \mu. \tag{12}$$

Here, Φ_e is the barrier height for electron emission, which according to Fig. 7(a) is equal to $\Phi_e^{(1)}$. At low temperatures, i.e., when $k_B T_e \ll \Phi_e$, Eq. (12) reduces to $\Phi'_e \approx \Phi$ and the sum in Eq. (11) reduces to the first term, yielding



FIG. 6. Calculated electron (squares), hole (circles), and total (stars) current density flowing from the top to the bottom layer of the Ag-AlO_x-Al junction. Following parameters have been used: $n_e^{Ag} = 5.86 \times 10^{28} \text{ m}^{-3}$, $\Phi_e = 3.0 \text{ eV}$, and $\Phi_h = 4.0 \text{ eV}$.

$$j_e = \frac{em}{2\pi^2\hbar^3} (k_B T_e)^2 e^{-\Phi_e/k_B T_e},$$
 (13)

which is the well-known Richardson-Dushman formula for thermionic emission.³³ At high temperatures, i.e., when $T_e \gg T_F$, the electron gas can be described by a Maxwell-Boltzmann distribution. Then, the chemical potential is given by Eq. (7) and all terms in Eq. (11) with n > 1 tend to zero, such that j_e reduces to

$$j_e \approx \frac{em}{2\pi^2 \hbar^3} (k_B T_e)^2 e^{\mu (T_e)/k_B T_e} \propto T_e^{1/2}.$$
 (14)

The hole current density can be derived in a similar way (see Appendix B) as

$$j_{h} = -\frac{em}{2\pi^{2}\hbar^{3}}k_{B}T_{e}\{-E_{0}\ln(e^{-\mu/k_{B}T_{e}}+1) + \int_{0}^{E_{0}}\ln[e^{(E-\mu)/k_{B}T_{e}}+1]dE\},$$
(15)

where $E_0 = E_F - \Phi_h$ defines the width of the energy band occupied by the holes emitted over the barrier Φ_h which, according to Fig. 7, is equal to $\Phi_h^{(2)}$. At low temperatures, i.e., when $T_e \ll T_F$, Eq. (15) reduces to

$$j_{h} = -\frac{em}{2\pi^{2}\hbar^{3}}(k_{B}T_{e})^{2}e^{-\Phi_{h}/k_{B}T_{e}}[1-(\eta+1)e^{-\eta}], \quad (16)$$

where $\eta = E_0/k_B T$. For an infinitely wide energy band of emitted holes, i.e., $E_0 \rightarrow \infty$, the exponent $e^{-\eta} \rightarrow 0$ and Eq. (16) reduce to the known Richardson-Dushman equation. Thus, the coefficient η introduces a correction to the Richardson-Dushman formula due to the finite width of the energy band of emitted holes. At high temperatures, i.e., when $T_e \gg T_F$, Eq. (15) reduces to

$$j_h \approx -\frac{em}{4\pi^2 \hbar^3} E_0^2 e^{-\beta\mu} \propto T_e^{3/2}.$$
 (17)

The calculated temperature dependences of the electron, hole, and total current density, respectively, is depicted in Fig. 6. First, one can observe that the hole contribution to the total tunneling current is small. Up to about 10^4 K, the total current density is determined by the Richardson-Dushman equation (13) and is negligibly small. The most important remark is, however, that the $j-T_e$ dependence can be well approximated by a straight line over a large temperature region.

The tunneling emission yield can be determined as

$$\gamma = (1/e)j\,\delta\!A\,\delta t\,,\tag{18}$$

where $\delta A \approx (\delta V)^{2/3}$ is the effective emission area. This relation predicts, within the model assumptions δV =const. and δt =const., that $\gamma \propto j \propto T_e$. By combining this with $T_e \propto E_{\text{pot}}$ given by relation (10), it further predicts that $\gamma \propto E_{\text{pot}}$, which agrees with the experimental data presented in Fig. 2. By differentiating Eq. (18) with respect to E_{pot} , we deduce

$$\frac{\partial \gamma}{\partial E_{\text{pot}}} = (1/e) \,\delta A \,\delta t \frac{\partial j}{\partial T_e} \frac{\partial T_e}{\partial E_{\text{pot}}}.$$
(19)

From Eq. (19), the slope $\partial T_e / \partial E_{pot}$ of the $T_e - E_{pot}$ dependence is estimated to 120 K/eV, where we assumed $\partial V = 448$ Å³, i.e., equal to the volume calculated in Sec. IV C, $\partial t = 10$ fs, $\partial j / \partial T_e$ is determined from the linear fit to the theoretical curve j - T in the energy region discussed here (dashed line in Fig. 6), and $\partial \gamma / \partial E_{pot}$ is determined from the linear fit to the experimental data $\gamma - E_{pot}$ (solid line in Fig. 2). The slope $\partial T_e / \partial E_{pot}$ estimated in this way is, hence, by about a factor of 2 smaller than that determined from Fig. 4 for the external emission from Cu.

Finally, in the high-temperature region $(T_e > 16 \times 10^4 \text{ K})$, *j* deviates slightly from this approximately linear behavior, tending to exhibit a rather square-root-like dependence on T_e , as predicted by Eq. (14). This is due to the fact that the electron gas becomes gradually nondegenerated.

Thus, the simple thermodynamical model presented here succeeds to explain, at least for charge states $q \leq 8$, the apparent linear $\gamma - E_{pot}$ dependence observed in both the internal and the external emission of electrons induced by Ar^{q+} ions at metal surfaces.^{14,15} For charge states q > 8, a strong deviation from this linearity was observed, ^{14,15,34} which does not seem to follow a square-root dependence as predicted by the present model. Kurz et al.¹⁴ explained this effect by the creation of L-shell vacancies which are not entirely being filled before the MCI reaches the surface. As a consequence, Auger transitions into the L shell become an important deexcitation channel at the surface, such that less electrons per unit of potential energy, but possessing higher kinetic energies, are emitted. This argument is confirmed, for example, by the intense Auger transition peak observed in the electron emission spectra from copper^{30,35} and tungsten,³⁴ at about 200 eV, when bombarded with Ar9+ ions. This peak can be interpreted as the result of the superposition of $L_{2,3}$ -MM Auger transitions lying in the energy range between 178 and 207 eV, as proved experimentally^{36,37} and theoretically.³⁸

In conclusion, the thermodynamic model presented in this work cannot be extended to argon charge states q > 8, since the contribution of nonthermalized Auger electrons to the external electron emission can no longer be neglected. With



FIG. 7. Energy scheme of a metal-insulator-metal tunnel junction: (a) without a bias voltage and (b) with a (positive) bias voltage U applied between the two metal electrodes (1) and (2). Nomenclature: (a) $\Phi_e^{(1)}$ and $\Phi_h^{(1)}$: barrier heights for electrons and holes, respectively, at the top-metal-oxide interface; $\Phi_e^{(2)}$, Φ_h^2 : barrier heights for electrons and holes, respectively, at the bottom-metal-oxide interface; E_g : band gap of the oxide; $E_F^{(1)}$, $E_F^{(2)}$: Fermi level of metal (1) and metal (2), respectively; (b) E: electron energy; $\Phi_e(z)$: electron barrier potential at a distance z from the top-metal-oxide interface; U: applied bias voltage; and d: thickness of the oxide layer.

increasing charge state, this contribution will dominate and the emission yield becomes rather proportional to the charge state q of the MCI.¹⁴

E. Yield-voltage dependence

By applying a bias voltage between the metal electrodes of the MIM junction, one modifies the potential barrier and, hence, the barrier heights Φ_e and Φ_h for electron and hole emission, respectively. To enable the use of the Eqs. (11) and (15), one has to introduce Φ_e and Φ_h as functions of the bias voltage U. In order to determine the bias dependence of the two barrier heights, we assume again an asymmetric trapezoidal energy barrier, as illustrated in Fig. 7(b). In addition, we took into account the effect of the image charge potential V_i which becomes important for thicknesses of the barrier in the nanometer range, as it is the case for the MIM junctions studied here. Φ_{e} is then determined by the maximum height of the tunnel barrier Φ_e^{max} , while Φ_h is given by the maximum depth Φ_h^{max} ; both taken relative to the Fermi level of the excited metal. The two extrema can be determined by finding the maximum relative to z of the barrier height functions

$$\Phi_e(U,z) = \Phi_e^{(1)} - (\Phi_e^{(1)} - \Phi_e^{(2)} + eU)\frac{z}{d} + V_i(z), \quad (20)$$

$$\Phi_h(U,z) = \Phi_h^{(1)} - (\Phi_h^{(1)} - \Phi_h^{(2)} - eU)\frac{z}{d} + V_i(z), \quad (21)$$

where z is the electron coordinate relative to the oxideexcited metal interface and d is the thickness of the barrier. The image charge potential $V_i(z)$ is defined as³⁹



FIG. 8. Calculated bias voltage dependence of the barrier heights Φ_e and Φ_h for electron and hole emission, respectively, for a trapezoidal barrier with $\Phi_e^{(1)} = \Phi_e^{(2)} = 3.0$ eV, $\Phi_h^{(1)} = \Phi_h^{(2)} = 4.0$ eV, d=4 nm, and $\epsilon_e = 8$.

$$V_i(z) = -\frac{e^2}{4\pi\epsilon_0\epsilon_r} \left[\frac{1}{2z} + \sum_{n=1}^{\infty} \left(\frac{nd}{(nd)^2 - z^2} - \frac{1}{nd}\right)\right].$$
 (22)

The extrema $\Phi_e^{\rm max}$ and $\Phi_h^{\rm max}$ are now determined by the conditions

$$\frac{d\Phi_e(U,z)}{dz} = -\left(\Phi_e^{(1)} - \Phi_e^{(2)} + eU\right)\frac{1}{d} + \frac{dV_i(z)}{dz} = 0, \quad (23)$$

$$\frac{d\Phi_h(U,z)}{dz} = -\left(\Phi_h^{(1)} - \Phi_e^{(2)} - eU\right)\frac{1}{d} + \frac{dV_i(z)}{dz} = 0.$$
 (24)

In order to simulate the experimental data in Fig. 3, we applied the present thermodynamical model to a rectangular, idealized MIM barrier structure, defined by the following parameters: $\Phi_e^{(1)} = \Phi_e^{(2)} = 3.0$ eV and $\Phi_h^{(1)} = \Phi_h^{(2)} = 4.0$ eV, d=4 nm and ϵ_r =8. Image charge effects are also included, leading to a rounding off of the corners and a lowering of the barrier height for electron and hole emission, respectively.³⁹ The corrected maximum barrier heights Φ_e^{\max} and Φ_h^{\max} are reduced by about 0.1 eV (at U=0 V) with respect to those corresponding to the idealized MIM structure and depend on bias voltage as illustrated in Fig. 8. It can be noticed that, with increasing positive bias voltage, Φ_e^{\max} drops slightly, while Φ_h^{max} increases apparently linearly. At negative voltages, Φ_e^{max} increases linearly with decreasing voltage, while Φ_{e}^{\max} is slightly reduced. Interestingly, the two barrier heights become equal at U=-1.0 V, this value corresponding exactly to $\Phi_e^{\max} - \Phi_h^{\max}$ at U=0 V.

Using the barrier heights calculated above, we calculated the j-U dependence for several electron temperatures. The results of these simulations are plotted in Fig. 9. At 10 000 K, the current shows the typical sign reversal already observed by us at low excitation energies¹⁷ and succeeds to explain qualitatively the experimental curve in Fig. 3, corresponding to a charge state q=1. The shape of the experimental curve is, however not well reproduced. One reason could be that the potential barrier is not as "rigid" as assumed by the theory. Thus, in a nonrigid barrier model, $\Phi_e^{(1)}$ decreases with increasing positive bias voltage, while it remains constant in the rigid barrier model. In the former case, the shape



FIG. 9. Total tunneling current density in a Ag-AlO_x-Al junction flowing from Ag into Al plotted as a function of the applied bias voltage at 1×10^4 K (circles), 3×10^4 K (triangles), and 15 $\times 10^4$ K (squares), respectively. The current is normalized to its value at 0 V. The following parameters have been used: n_e^{Ag} =5.86 $\times 10^{28}$ m⁻³, Φ_e =3.0 eV, and Φ_b =4.0 eV.

of the calculated curve at 10 000 K (not shown here) was found to approach the experimental curve plotted for q=1. While at 10 000 K, the sign reversal occurs at U=-1.25 eV, it shifts with increasing temperature to higher (negative) values, such that, at 30 000 K, no sign reversal is observed in the investigated voltage range. At very high temperatures, i.e., at 150 000 K, the i-U curve becomes nearly flat (see Fig. 9), that is, the tunneling current is only slightly influenced by the bias voltage. This temperature dependence can be interpreted as follows: At lower temperatures, the bias voltage, where the sign reversal of the tunneling yield occurs, is primarily given by the difference between the barrier heights for electron and hole emission, respectively.¹⁷ This is due to the symmetry of the electron-hole energy distribution relative to the chemical-potential level, which is approximately situated at the same energy as the Fermi level. At higher temperatures, the chemical potential shifts downwards, i.e., toward the bottom of the conduction band of the electron gas. Consequently, the symmetry relative to the Fermi level breaks down. On the other hand, the energy band of excited holes is limited to the region between 0 and E_F $[E_F=5.5 \text{ eV for Ag (Ref. 25)}]$. As a result, the increase in electron emission can be only partially compensated by the hole emission. Above 150 000 K, j_h seems to saturate, while j_e further increases (see Fig. 6), such that the relative contribution of the holes becomes negligible. A second reason for the observed flatness of the j-U curve at 150 000 K is the increased production of high energy electrons, which are hardly influenced by the bias voltage. Experimentally, the tunneling yield induced by Ar⁸⁺ shows a similar behavior, varying by less then 10% in the voltage range from -1.0 to 1.0 eV (see Fig. 3). This result nicely confirms the high value of the electron temperature of about 150 000 K derived from Ref. 30 for the $Ar^{\hat{8}+}$ induced emission from copper.

V. CONCLUSIONS

In the present paper, we presented a study of the internal electron emission induced by multiply charged argon ions in Ag-AlO_x-Al thin film tunnel junctions. Experimentally, we

observed an apparently linear dependence of the induced tunneling yield on the total potential energy of the MCI. Such behavior was already observed in the case of the external emission yield, too. We introduced an "electron temperature" within a thermodynamical free-electron model to describe the energy distribution of the emitted electrons and holes. Despite its simplicity, the model succeeds to explain the observed linear dependence of both the internal and external emission yields on the potential energy up to a charge state q=8 of the argon MCI. For $q \ge 9$ our model cannot be applied, since the external electron emission was shown to be dominated by nonthermalized Auger electrons. Valuable information on the energy distribution of the excited charge carriers can be gathered from the experimentally determined bias voltage dependence of the tunneling yield. The yieldvoltage dependence indicates that metal-insulator-metal junctions may act as sensors for electron temperatures. Thus, it is shown that the hotter the excited electrons, the weaker the influence of the bias voltage on the emission yield. Even though the simple thermodynamic model presented here seems to be able to explain the experimental results on external and internal emissions induced by multiply charged ions, a more evolved theory and further experiments with variable thickness of the top-metal film are needed in order to understand the mechanisms of internal electron emission. The theory should include ion penetration (nuclear and electronic stoppings) and the transport of hot charge carriers across the top metal of such MIM junctions (elastic and inelastic scatterings). Such investigations are under way.

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APPENDIX A

The current density of electrons escaping from the metal is given by

$$j_e = e \cdot \int \frac{d\mathbf{k}}{4\pi^3} v_x f(\mathbf{k}), \qquad (A1)$$

where

$$v_x = \frac{\hbar k_x}{m} \tag{A2}$$

is the electron velocity in vacuum,

$$f(\mathbf{k}) = \frac{1}{\exp\{\beta[E(\mathbf{k}) - \mu]\} + 1}$$
(A3)

is the Fermi-Dirac distribution function of the emitted electrons, and **k** is the wave vector outside the metal. In Eq. (A2), k_x is the component of the wave vector perpendicular

to the surface and *m* is the mass of the electron. In Eq. (A3), $E(\mathbf{k})$ is the electron energy defined outside of the metal as

$$E(\mathbf{k}) = \frac{\hbar^2 k^2}{2m} + E_F + \Phi_e, \qquad (A4)$$

with E_F being the Fermi energy and Φ_e the barrier height for electrons. Substituting Eq. (A4) in Eq. (A3) and introducing the notation

$$\Phi'_e = \Phi_e + E_F - \mu, \tag{A5}$$

Equation (A3) can be rewritten as

$$f(\mathbf{k}) = \frac{1}{\exp\left[\frac{1}{k_B T} \left(\frac{\hbar^2 k^2}{2m} + \Phi'_e\right)\right] + 1}.$$
 (A6)

Changing to spherical coordinates, j_e becomes

$$j_e = \frac{e\hbar}{4\pi^2 m} \int_0^\infty \left[e^{\beta(\hbar^2 k^2/2m + \Phi'_e)} + 1 \right]^{-1} k^3 dk.$$
 (A7)

Making the substitution $k \rightarrow E$ according to Eq. (A4), one obtains

$$j_e = \frac{em}{2\pi^2\hbar^3} \int_{E_F + \Phi_e}^{\infty} (E - E_F - \Phi_e) [e^{\beta(E-\mu)} + 1]^{-1} dE.$$
(A8)

By using the property

$$(e^{ax+b}+1)^{-1} = -\frac{1}{a}\frac{d}{dx}\ln[e^{-(ax+b)}+1],$$
 (A9)

Eq. (A8) can be integrated by parts, yielding

$$j_{e} = \frac{em}{2\beta\pi^{2}\hbar^{3}} \int_{E_{F}+\Phi_{e}}^{\infty} \ln[e^{-\beta(E-\mu)} + 1] dE.$$
 (A10)

Since $e^{-\beta(E-\mu)} \le 1$ over the whole integration region, one can expand the logarithmic function in a Taylor series according to

$$\ln(1+x) = \sum_{n=1}^{\infty} (-1)^{n-1} \frac{x^n}{n}, \quad -1 < x \le 1.$$
 (A11)

Integrating then each element of the Taylor series, the electron current density takes the form

$$j_e = \frac{em}{2\pi^2\hbar^3} (k_B T_e)^2 \sum_{n=1}^{\infty} (-1)^{n-1} \frac{1}{n^2} e^{-n\Phi_e'/k_B T_e}.$$
 (A12)

APPENDIX B

The hole current density j_h escaping from the top metal can be introduced in a similar way as the electron current density j_e , namely

$$j_h = -e \cdot \int \frac{d\mathbf{k}}{4\pi^3} v_x [1 - f(\mathbf{k})], \qquad (B1)$$

where $1-f(\mathbf{k})$ is the hole distribution function, while v_x and $f(\mathbf{k})$ are defined as in Eqs. (A2) and (A3), respectively. The

dispersion relation $E(\mathbf{k})$ in Eq. (A4) has to be modified, however, according to

$$E(\mathbf{k}) = -\frac{\hbar^2 k^2}{2m} + E_F - \Phi_h, \qquad (B2)$$

where Φ_h is the barrier height for holes. Substituting Eq. (B2) in Eq. (B1) and introducing the notation

$$\Phi_h' = \Phi_h - (E_F - \mu), \tag{B3}$$

the hole distribution function can be written as

$$1 - f(\mathbf{k}) = \left[e^{\beta(\hbar^2 k^2/2m + \Phi'_h)} + 1\right]^{-1},$$
 (B4)

which is formally identical with Eq. (A6). Substituting Eqs. (A2) and (B4) in Eq. (B1) and observing that the integration is limited by the conditions $E \ge 0$ and $k_x > 0$ to a hemisphere S_k in the **k** space, with the radius

$$k_0 = \frac{1}{\hbar} \sqrt{2m(E_F - \Phi_h)},\tag{B5}$$

one obtains

$$j_{h} = -\frac{e\hbar}{4\pi^{3}m} \int_{S_{k}} \left[e^{\beta(\hbar^{2}k^{2}/2m + \Phi_{h}')} + 1 \right]^{-1} k_{x} d\mathbf{k}.$$
 (B6)

Changing to spherical coordinates, j_h becomes

$$j_{h} = -\frac{e\hbar}{4\pi^{2}m} \int_{0}^{k_{0}} \left[e^{\beta(\hbar^{2}k^{2}/2m + \Phi_{h}')} + 1 \right]^{-1} k^{3} dk.$$
 (B7)

By substituting E for k according to Eq. (B2) and taking into account Eq. (B5), Eq. (B7) takes the form

$$j_h = -\frac{em}{2\pi^2\hbar^3} \int_0^{E_0} \left[e^{\beta(\mu-E)} + 1 \right]^{-1} (E_0 - E) dE, \qquad (B8)$$

with $E_0 = \hbar^2 k_0^2 / 2m$. By integrating by parts, this can be expanded to

$$j_{h} = -\frac{em}{2\beta\pi^{2}\hbar^{3}} \{ -E_{0} \ln(e^{-\beta\mu} + 1) + \int_{0}^{E_{0}} \ln[e^{\beta(E-\mu)} + 1] dE \},$$
(B9)

which, again, cannot be solved analytically. For solving the equation numerically, it is convenient to expand the function under the integral in a Taylor series. Here, however, one needs to distinguish three cases:

(a) $\mu \ge E_0$. This situation is met at low temperatures, i.e., when $T_e \ll T_F$. In this case, Eq. (A11) can be applied, yielding

$$j_{h} = -\frac{em}{2\beta^{2}\pi^{2}\hbar^{3}} \left[\sum_{n=1}^{\infty} (-1)^{n-1} \frac{1}{n^{2}} e^{-n\beta\Phi_{h}'} (1 - e^{-n\beta E_{0}}) -\beta\epsilon_{0} \ln(e^{-\beta\mu} + 1) \right].$$
(B10)

For the particular case that $k_B T_e \ll \Phi_h$, $\Phi'_h \approx \Phi_h$ and the sum in Eq. (B10) reduce to the first term, leading to

$$j_{h} = -\frac{em}{2\pi^{2}\hbar^{3}}(k_{B}T_{e})^{2}e^{-\Phi_{h}/k_{B}T_{e}}[1-(\eta+1)e^{-\eta}], \quad (B11)$$

where $\eta = E_0 / k_B T_e$.

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(b) $0 < \mu < E_0$. In order to apply the Taylor series, the integral in Eq. (B9) must be written as

$$\int_{0}^{\mu} \ln[e^{\beta(E-\mu)} + 1]dE + \int_{\mu}^{E_{0}} \ln[e^{-\beta(E-\mu)} + 1]dE + \beta(E_{0} - \mu).$$
(B12)

Applying Eq. (A11) to both integrals, Eq. (B9) becomes

$$j_{h} = -\frac{em}{2\beta^{2}\pi^{2}\hbar^{3}} \left[\frac{\pi^{2}}{6} - \beta\Phi_{h}' - \beta E_{0} \ln(e^{-\beta\mu} + 1) - \sum_{n=1}^{\infty} (-1)^{n-1} \frac{1}{n^{2}} (e^{n\beta\Phi_{h}'} + e^{-n\beta\mu}) \right],$$
 (B13)

where the property

$$\sum_{n=1}^{\infty} (-1)^{n-1} \frac{1}{n^2} = \frac{\pi^2}{12}$$
(B14)

was used.

(c) $\mu \leq 0$. The integrand in Eq. (B9) can be expanded in a Taylor series if rewriting it as

$$\ln[e^{\beta(E-\mu)} + 1] = \ln[e^{-\beta(E-\mu)} + 1] + \beta(E-\mu). \quad (B15)$$

Then, Eq. (B9) results in

$$j_{h} = -\frac{em}{2\beta^{2}\pi^{2}\hbar^{3}} \left[\sum_{n=1}^{\infty} (-1)^{n-1} \frac{1}{n^{2}} (e^{n\beta\mu} - e^{n\beta\Phi'_{h}}) + \beta E_{0} - \beta E_{0} \ln(e^{-\beta\mu} + 1) \right].$$
(B16)

At temperatures $T_e \gg T_F$, due to $\beta \mu \ll 0$, one obtains

$$j_h \approx -\frac{em}{4\pi^2\hbar^3} E_0^2 e^{-\beta\mu} \propto T_e^{3/2}.$$
 (B17)

*detlef.diesing@uni-due.de

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