$\hbar^2)^2$ and equals 1.1 meV. For $E = 8$ meV Eq. (6) gives $\rho = 7\times 10^{15}$ states/meV cm^3. This is close to the value of ρ we deduced from the data under the assumption of equal quantum efficiencies for D_0^X and M_0^X.

We have assumed nonzero short-range phosphorus atom correlations. They should be looked for by x-ray diffraction techniques.

We believe that the most reasonable explanation of the M_0^X line is that is is due to recombination of alloy-trapped excitons. More detailed experimental and theoretical results will be forthcoming.

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[a]Present address: Physics Department, University of Wisconsin, Madison, Wisc. 53706.

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Plasmon Gains as a Monitor of Incomplete Relaxation, Interference Effects, and the Transition from Sudden to Adiabatic Limits in Electron Spectroscopies

John C. Fuggle and Rainer Lässer[a]
Institut für Festkörperforschung der Kernforschungsanlage, D-5170 Jülich, West Germany

and

Olle Gunnarsson
Max-Planck-Institut für Festkörperforschung, Busnau, D-7000 Stuttgart, West Germany

and

Kurt Schönhammer
Institut für Theoretische Physik der Universität Hamburg, D-2000 Hamburg 36, West Germany

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This Letter reports on the *KLL* Auger spectra of Na and Mg excited with different x-ray sources. Intrinsic plasmon creation in x-ray photoemission leads to plasmon-gain satellites in the Auger process. With low-energy x rays the probability of intrinsic plasmon creation in x-ray photoemission and hence the plasmon-gain probability is reduced. With high-energy x rays the plasmon-gain intensities agree with predictions of a new formalism of photoemission and Auger transitions as a single quantum mechanical process.

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It is the purpose of this paper to show that plasmon-gain processes in Auger electron spectroscopy (AES) present new possibilities for investigating phenomena of incomplete relaxstion, interference effects, and the changeover from the adiabatic to the sudden limits in photoelectron

spectroscopy. We believe that this is the first quantitative experimental determination of the variation with photon energy of the intrinsic plasmon creation probability in x-ray photoemission spectroscopy (XPS) and of the effects of adiabatic-to-sudden changeover on shakeup.[1] A new one-step model of the XPS and Auger processes successfully gives the intensities of the plasmon gain in Na, Mg, and Al when high-energy x-ray excitation is used.

In the theory of photoemission it is predicted that shakeup processes should be weak when the photoelectron kinetic energy is low and the excitation is adiabatic. A good test of the theoretical arguments should be provided by experiments on free-electron-like metals such as Na and Mg, where the shakeup intensity appears mainly as plasmon satellites. Here the transition from sudden to adiabatic excitation should take the form of interference between the *intrinsic* shakeup and *extrinsic* inelastic losses.[2-4] Experimental evidence confirming these arguments is, however, scarce. The intrinsic and extrinsic plasmons can be separated by use of the intensities of multiple losses, but such attempts have led to varying and hence questionable results.[5-8]

In the usual two-step model of Auger emission[9] it is assumed that intrinsic plasmons excited in the XPS process decay or propagate away before Auger decay of the core hole. However, plasmon lifetimes in Na (0.4 eV^{-1}) and Mg (0.7 eV^{-1}) (Ref. 10) are comparable to K hole lifetimes (0.30 and 0.36 eV^{-1}, respectively.[11] Thus simultaneous Auger decay of the core hole and plasmons can occur and give rise to a high-energy plasmon-gain satellite.[12-17] This is a classic example of incomplete relaxation. Extrinsic plasmons contribute only weakly, or not at all, because they have small concentration at the core ionized atom. As factors determining the plasmon-gain intensity such as the ratio of core-hole and plasmon lifetimes and plasmon dispersions are all constant for a given metal, we can use the variation with photon energy of the relative plasmon-gain intensity, β^+, to investigate the changeover from adiabatic to sudden limits in XPS. That is, we can assume that the plasmon-gain probability, β^+, in AES is proportional to the intrinsic plasmon loss probability, β^-, in XPS.

A combination of Mg, Al, Si, Ti, and Cu anodes with Al or Be window materials was used to obtain either almost pure Mg $K\alpha$ (1253.6 eV) or Al $K\alpha$ (1486.7 eV) radiation or pure bremsstrahlung radiation with an average energy of about 10 keV. In addition, a mixture of Si $K\alpha$ (1740 eV) and bremsstrahlung radiation could be produced and contributions of these two sorts of radiation to the XPS and Auger spectra were separated arithmetically. The energies of the Na KLL and Mg KLL Auger peaks used in this study are ~ 995 and ~ 1186 eV, respectively.

Figure 1 illustrates the Na $KL_{2,3}L_{2,3}$ ^1D Auger spectrum excited with Mg $K\alpha$ radiation together with the plasmon gain and the first plasmon loss. The mere existence of the plasmon-gain peak shows that there is an intrinsic contribution to the plasmon loss found in XPS and that it is important to use a one-step model of XPS and AES.[12-17] The figure also shows a magnified plot of the plasmon-gain peak excited with Mg $K\alpha$ and bremsstrahlung radiation with energies of 1253.6 and ~ 10 000 eV, respectively, and normalized with respect to the Na ^1D peak. The intensities of the plasmon-gain peak and of the region between the gain and the main peaks clearly increase with the energy of the exciting radiation.[18] This reflects the increased intrinsic plasmon production in the XPS process with increasing photoelectron kinetic energy, E. This variation in the intrinsic plasmon-loss coupling strength, β_E^-, with E due to interference with the extrinsic process has been expressed by Gadzuk[4] as

$$\beta_E^- = \beta_\infty^- - (e^2/\hbar)F(q_c\nu/\omega_p), \quad (1)$$

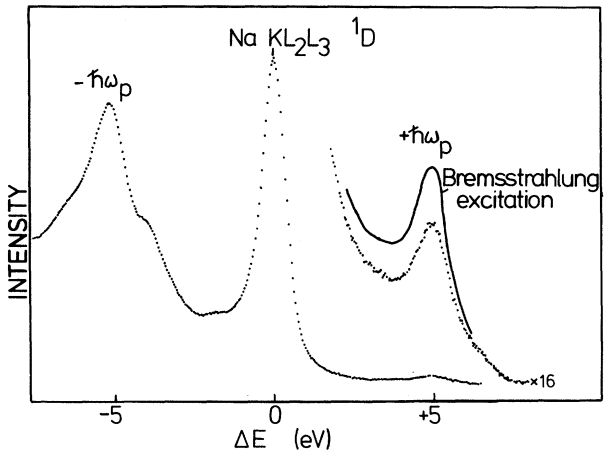

FIG. 1. Na KL_2L_3 ^1D Auger peak, with associated plasmon loss and gain, as excited by Mg $K\alpha$ radiation. Both surface and bulk plasmon losses are observed and some intensity ~ 5 eV below the main peak is due to the Na KL_2L_2 ^1S peak. Also shown, with the intensity scale expanded, is the gain region as excited with Mg $K\alpha$ radiation and with bremsstrahlung radiation (solid line). The spectra have been normalized for the KL_2L_3 ^1D peak intensities.

where β_∞^- is the coupling constant without interference, i.e., for infinite photoelectron energy and $F(x)$ is a slowly varying function.

In Fig. 2 we have plotted β_E^+/β_{br}^+, as a function of the photoelectron energy, where β_{br}^+ was obtained for bremsstrahlung radiation with a peak intensity at ~10 keV. Thus β_{br}^- is only about 10% smaller than β_∞^- according to theory.[19] Experimentally, the ratio β_E^+/β_{br}^+ was found for Na KLL ^1D with Mg $K\alpha$, Al $K\alpha$, and Si $K\alpha$ radiation, and for Mg KLL ^1D with Al $K\alpha$ and Si $K\alpha$ radiation. Comparison with Gadzuk's approximate calculation shows an overall agreement, although theory seems to overestimate the importance of interference effects as does the newer formalism of Chastenet and Longe.[20] A different treatment of the sudden-to-adiabatic transition for valence-level photoemission by Schrieffer[21] produces a changeover to adiabatic behavior at much lower photoelectron kinetic energy, so that our experimental approach should provide a useful guide to the applicability of the different theories. The possibility that "intrinsic" plasmon gains play a small role when photoelectron kinetic energies are low should be investigated because inelastic mean free paths are then very short.

Using the sudden approximation, we have developed a theory where the XPS and Auger processes are treated in a unified manner. The Auger decay is included to all orders and the finite plasmon lifetime is taken into account.[22] In the region of the plasmon gain peak the Auger electron current is given by

$$j(\epsilon) \sim \left| \frac{1}{\epsilon - i\Gamma} - \sum_q \left(\frac{g_q}{\omega_q}\right)^2 \frac{1}{\epsilon - \omega_q - i\Gamma_{pl} - i\Gamma} \right|^2, \quad (2)$$

where 2Γ and $2\Gamma_{pl}$ are the lifetime broadenings (full widths at half maximum) of the core level and plasmon, respectively. ω_q is the plasmon energy at momentum q and the coupling constant between the core hole and the plasmon is given by g_q.[23] The main peak and the plasmon gain satellite are given by the squares of the first and second terms, respectively. The cross term represents an interference between these two processes.

In Table I we list the experimental heights of the Na, Mg, and Al plasmon gain peaks relative to the ^1D KLL peaks in bremsstrahlung-excited Auger spectra. All possible disturbing effects of plasmon losses from the KLV Auger peaks or the primary peak have been subtracted.[7,24] Agreement between theory and experiment is poor if $2\Gamma_{pl}$ is set equal to 0 in Eq. (2) as if the intrinsic plasmons did not decay at all prior to Auger decay of the core holes. However, inclusion of the appropriate value[25] of $2\Gamma_{pl}$ gives very good agreement considering the experimental accuracy of the parameters needed to evaluate Eq. (2).[10] The agreement would be improved if the slight deviation from the sudden limit, even with bremsstrahlung radiation, were included. It must also be remembered that emission from the surface atoms contributes to the primary peak, but not to the bulk-plasmon-gain peak, thus decreasing the value of β^+ by about 20% in our experiments.

In summary, our results indicate that in Auger spectroscopy the ratio of the core-hole and shake-up-excitation lifetimes is an important factor in determining the strength of incomplete relaxation effects. Interference between intrinsic and extrinsic losses have been shown experimentally to reduce the intrinsic loss contribution to photoelectron spectra by ~40% when the photoelectron energy is ~20 times the loss energy and the change-

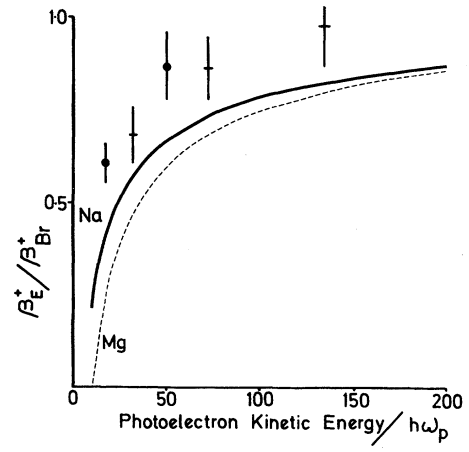

FIG. 2. The decrease of the plasmon gain intensity, β^+, relative to the main KL_2L_3 ^1D Auger peak due to onset of adiabatic effects in the XPS process at low photoelectron energy for Na(plusses) and Mg(dots). Calculation of theoretical curves for Na(solid line) and Mg(dashed line) is described in the text.

TABLE I. Heights of experimental and theoretical plasmon gain peaks with respect to the main KLL ^1D Auger peak.

	Expt.	Theory, $2\Gamma_{pl} = 0$	Theory with $2\Gamma_{pl}$ from Ref. 25
Na	0.030 ± 0.003	0.067	0.032
Mg	0.0065 ± 0.002	0.024	0.0086
Al	0.005 ± 0.002	0.016	0.007

over from sudden to adiabatic excitation is continuous. No theory available at the present time gives a fully accurate description of the change-over as observed by studies of plasmon gains.

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[a]Present address: Bell Laboratories, Murray Hill, N. J. 07974.

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